UNIVERSITY OF LATVIA
DEPARTMENT OF PHYSICS AND MATHEMATICS

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Laser Ablation Spectroscopy for Impurity Depth Profiling in Hot Wall Materials of Thermonuclear Fusion Reactors

Summary of the Doctoral Thesis

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The defence of the PhD thesis will take place in an open session of the Promotion Council of Physics and Astronomy of the University of Latvia on the 27th of November, 2009, at 15:00 in the conference hall of the Institute of Solid State Physics of the University of Latvia.

The PhD thesis and its summary are available at the Library of the University of Latvia (4 Kalpaka Blvd, Rīga) and Latvian Academic Library (10 Rūpniecības St, Rīga).

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Contents

Abstract

The main goal of this study was to show the efficiency of laser-induced ablation in analysis of the impurities in plasma facing components, as the method allows to detect any chemical element and can be employed for remote analysis.

The major tasks of the present investigation are: to set up and test the equipment for laser ablation spectroscopy and to develop a methodology for impurity depth profiling.

The thesis describes the investigation of plasma facing materials of ASDEX Upgrade tokamak by means of laser-induced ablation spectroscopy and profilometry of the corresponding ablation craters.

The experimental setup for laser-induced ablation spectroscopy was developed and partly manufactured at the Institute of Solid State Physics. The optimal conditions of laser-induced ablation of the samples of ASDEX Upgrade divertor plates were discovered. Plasma emission spectra of these plasma facing components showing a substantial number of impurities were recorded. The impurity elements were determined, and the possible sources of the impurities suggested. The depth of the accumulation of basic impurities (hydrogen, boron) in the surface of the ASDEX Upgrade divertor plates (carbon R6710 tiles) was estimated using plasma emission spectra.

The obtained results allow to consider the method of laser-induced ablation spectroscopy to be feasible to carry out rapid analysis of plasma facing materials. With minor modifications, this method can be suggested as an *in situ* technique for determination of the state of the plasma facing components inside the chamber of a thermonuclear fusion reactor.

The Goal of the Doctoral Thesis

The goal of the thesis is to investigate plasma facing materials of the ASDEX Upgrade tokamak using the method of laser ablation spectroscopy that shows the impurity content in a given plasma facing component, as well as the depth of the accumulation of the impurities of major concern – i.e., hydrogen and boron.

The Tasks of the Doctoral Thesis

The major tasks of the thesis are the following:

- to set up and test the experimental equipment for laser-induced breakdown spectroscopy analysis of plasma facing materials;
- to develop a methodology for laser ablation of impurity depth profiling;
- to prove that laser-induced ablation spectroscopy is a feasible technique for rapid analysis of plasma facing materials with a prospect to develop an *in situ* method of inspection of the status of plasma facing materials inside the tokamak.

1 Introduction

The main issue in the design and construction of a thermonuclear fusion reactor with magnetically confined plasma is the interaction of the hot plasma with the material components of such a device. Plasma facing components of a vessel act as a reservoir for energy and particles coming from the plasma. At the same time, the interaction of particles with the surface of the material may cause the wall atoms and the atoms implanted from the fuel to release and enter the plasma.

The object of the investigation was a plasma facing component of the ASDEX Upgrade tokamak – carbon R6710 divertor tiles. An investigation of the content and the accumulation depth of the impurities implanted in this material was carried out.

1.1 The Research Status of the Subject

The optimal plasma facing material for future fusion devices has not been found yet. The choice of materials for plasma facing components in fusion reactors is determined by the interaction of plasma with these materials. The most important aspects are the lifespan of plasma facing components and the deuterium/tritium inventory problem. Various surface diagnostic methods are employed to study the processes caused by the plasma-material interaction, such as ion beam techniques [1, 2], ion probes [3], X-ray photoelectron spectroscopy [4], and many others.

In comparison to the mentioned techniques, there are certain advantages to laser-induced ablation spectroscopy. It is a virtually non-destructive analysis since only a tiny amount of material is consumed during the process. The method of laser ablation spectroscopy allows to perform elemental analysis of any material irrespective of its physical state. Any chemical elements can be detected.

1.2 The Motivation for the Thesis

The main motivation for this study was to show the efficiency of laser-induced ablation in analysis of impurities in plasma facing components, as the method allows to detect any chemical elements and can be employed for remote analysis. This is an essential issue with plasma facing materials because the detection of hydrogen isotopes and detritiation are among the crucial tasks of the analysis of the surface of plasma facing components, and because their irregular shape form spaces which are difficult to reach.

The goal of the study was to investigate plasma facing materials of the ASDEX Upgrade tokamak using the method of laser ablation spectroscopy that shows the impurity content in a given plasma facing component, as well as the depth of the accumulation of the impurities of major concern – i.e., hydrogen and boron.

The major tasks of the investigation are the following:

- setting up and testing the experimental equipment for the laser induced breakdown spectroscopy analysis of the plasma facing materials;
- developing the methodology of the laser ablation of impurity depth profiling;
- proving that the laser-induced ablation spectroscopy is a feasible technique for the rapid analysis of plasma facing materials with a perspective to develop *in situ* method of inspection the status of plasma facing materials inside the tokamak.

1.3 The Author's Contribution

This study was performed in co-operation with several scientific collaborators. The author's contribution was the setup and maintenance of the equipment for the experiment, making measurements, processing and summarizing of data. The vacuum chamber used in the investigations was designed by the engineer Jānis Straumēns. The contribution of Anatolijs Sarakovskis was the help with the experimental setup and design of the experiment. The contribution of Boris Polyakov was valuable discussions and practical help. In turn, the author's contribution to the publication "Some aspects of pulsed laser deposition of Si nanocrystalline films" by B. Polyakov et al. was the assistance with the procedure of the experiment and discussions.

The results of this study were discussed at the scientific seminar held on 17 September 2008 at the Institute of Solid State Physics, the University of Latvia. The results were published in 5 articles and presented at 14 conferences.

2 Experimental Techniques

2.1 Target Materials

Two kinds of ablation materials were used in this research. For reference measurements, R6710 graphite was used. The samples drilled out of the ASDEX Upgrade (AUG) tokamak divertor tiles after they were subjected to plasma discharges were the next material used in this study. Figure 2.1 shows both unablated and ablated samples.

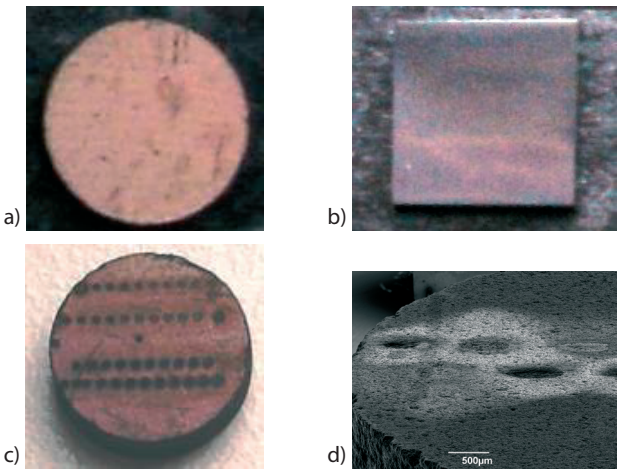

Fig. 2.1. **Samples used in this thesis: a) unablated ASDEX Upgrade target; b) unablated graphite R6710 sample; c) ablated ASDEX Upgrade target; d) a SEM image of an ablated ASDEX Upgrade target**

The concentrations of deuterium and boron in the samples cut from the divertor plates of ASDEX Upgrade were measured by means of Rutherford backscattering spectroscopy at the *Max-Planck-Institut für Plasmaphysic*, Garching.

2.2 Experimental Setup

The scheme of the experimental setup used in this research is shown in Figure 2.2. It consists of an Nd : YAG laser equipped with an optical system that steers and focuses the laser beam, a vacuum chamber, a pump system, and a spectroscopic system.

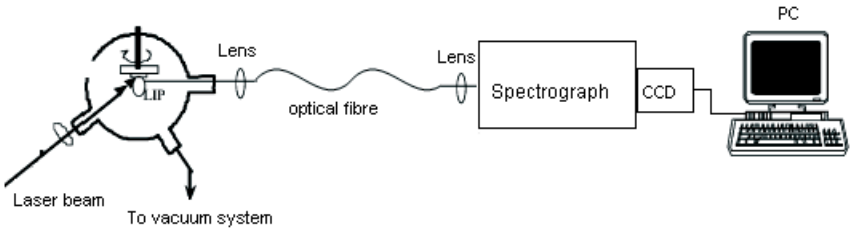

Fig. 2.2. **The scheme of the experimental setup**

The energy source of material evaporation and atomization in a pulsed laser ablation experiment is a laser. The cascade breakdown threshold scales as λ^{-2} [5, 6], which means that the minimal laser intensity required to achieve a breakdown in metal vapour or in ambient gas is easier to obtain with an IR laser than with UV. However, the ablation rate of mass is larger using an UV laser [7]. As the experiment requires ablation of the minimal possible mass, an infrared laser (1064 nm) was chosen.

As the laser-induced plasma source, a Q-switched Nd : YAG laser (SL-312, EKSPLA) with an emission wavelength of 1064 nm was used. It operates at a pulse repetition rate of 10 Hz, pulse width of 135 ps, and tuneable pulse energy up to 250 mJ [8]. To avoid spark ignition in the air, the beam was slightly defocused [9]. A convex quartz lens focused the beam at a distance of 3 mm inside the sample which was fixed either on an XYZ translation stage (measurements at atmospheric pressure) or inside the vacuum chamber. The average energy of the laser beam was monitored with a power/energy meter (Ophir, Model PE25BB-DIF). A quartz lens projected the plume image of the laser-induced plasma onto the entrance of the optical fibre which was placed in the image plane of the plume.

The emission light from the output end of the fibre bundle was collected to the slit of Andor Shamrock sr-303i spectrograph with a grating of 1200 gr/mm using a quartz lens of a focal distance of 50 mm. The spectrograph was equipped with an Andor CCD camera. The spectrograph was calibrated by a Hg lamp using a Hg yellow doublet (5789.7 and 5769.6 Å mercury lines).

To identify the elements of the spectra obtained in experiments, tabulated values of these elements were used. Integrating the intensity of carbon spectral lines, the uncertainty of the measurements did not exceed 10%. The uncertainties of other elements have to be estimated with reference to carbon spectral lines.

The depth of the ablation craters was estimated with Dektak 150 Surface Profiler (Veeco Instruments, Inc.). At the regime used for estimating the depth of the ablation craters, the profilometer provided a vertical bit resolution down

to 100 nm. The uncertainty of the measurements was mostly determined by the porous structure of the material.

2.3. The Vacuum Chamber

The vacuum chamber used in these experiments was constructed and manufactured at the workshops of the Institute of Solid State Physics. The vacuum chamber is shown in Figure 2.3. The positioning device inside the chamber allows the sample to be adjusted horizontally. Rotary motion feedthrough provides rotation of the sample, so that a series of experiments can be carried out in the same conditions. All the viewports are equipped with quartz windows. The entrance port for the ablation beam is designed to be sufficiently longer than the others in order to protect the viewport from contamination with the particles coming from the plasma plume (ablation is performed normally to the sample surface). A laser beam was delivered to the chamber through the lens of a focal distance of 300 mm. During the ablation, some material was unavoidably deposited on the windows, therefore they were cleaned when necessary.

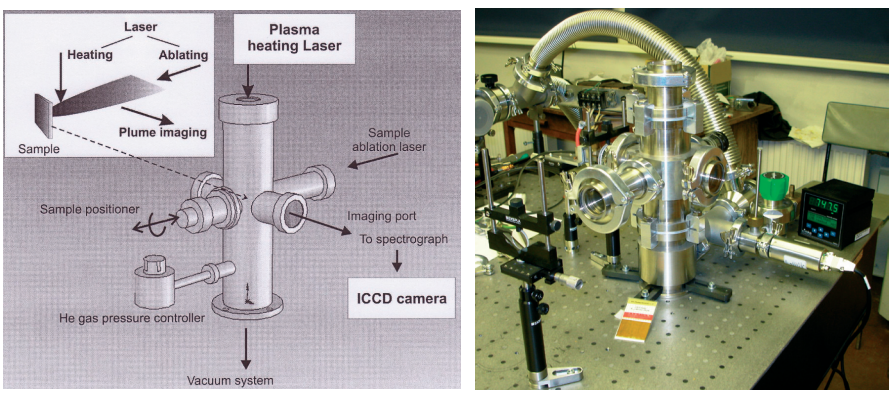

Fig. 2.3. **The vacuum chamber used in the experiments**

The development of the vacuum chamber allows to modify the system easily so that the second laser for a double-pulse experiment can be attached. Plasma can be heated from the upper viewport.

The chamber is connected to a turbomolecular pump (CDK 280 Ilmvac) providing a vacuum up to 10^{-5} Torr. The pumping system can be isolated from the vacuum chamber via an angle valve. The vacuum system is equipped with a pressure controller Series 910 DualTrans MicroPirani Absolute Piezo Transducer (MKS Instruments).

3 The Main Results

3.1 Ablation in Air/Atmospheric Pressure

To investigate the content of the impurities of the ASDEX Upgrade sample taken from the divertor region of the tokamak, laser-induced plasma emission spectra were recorded. An unexposed graphite R7610 sample was used as a reference sample. This sample contained no impurities typical of the tokamak sample. To get the profiles of the characteristic spectral lines, 10 laser pulses of energy of 10 mJ were applied to both samples [10]. The spectra are represented in Fig. 3.1.

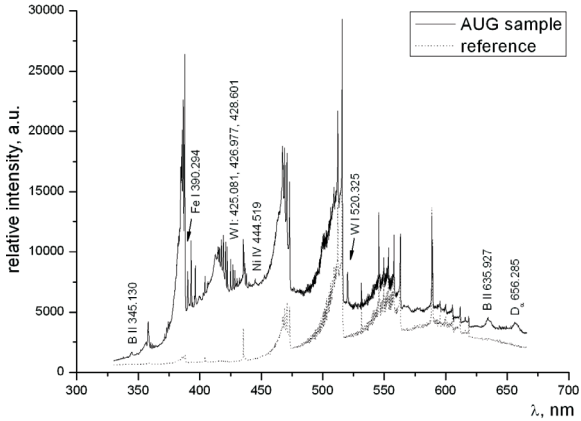

Fig. 3.1. **Laser-induced plasma emission spectra of graphite R7610 reference sample and ASDEX Upgrade sample. Carbon lines are not specified in the figure [11]**

The spectrum of the ASDEX Upgrade sample shows a substantial number of impurities. Tungsten lines (W I 425.061 nm, 426.977 nm, and 428.601 nm) are detected since tungsten is the main material for the first wall in ASDEX Upgrade [12]. The most probable reason for the appearance of B II (345.130 nm, 634.927 nm) lines might be a process of boronization of plasma facing components – a procedure of surface conditioning [13]. Surface conditioning is performed to optimize the influence of plasma facing components on the plasma performance. An unconditioned wall releases gaseous species, which can prevent stable plasma operation. In order to minimize the gas release, surface conditioning reduces the wall inventory [14].

The source of Fe I 390.294 nm and Ni IV 444.519 nm might be stainless steel components which are not shielded from plasma, mainly at the outer wall of the main chamber and, in addition, the outer covering of electric cables [15]. These cables are moving during discharges, and this movement is the reason for crack formation. Release of material from these cracks may lead to formation of dust

[16, 17] which migrates through the machine and is afterwards re-deposited in various places.

Although both spectra were recorded under the same conditions, the relative intensities of the spectral lines belonging to the reference sample were considerably lower than those of the ASDEX Upgrade sample. This might be due to surface differences in the ablated samples (see Fig. 2.1a, b). Reference graphite R7610 sample was polished smoothly in contrast to the ASDEX Upgrade sample, which is much rougher and exhibits traces of erosion after the plasma discharge. The ablation rate of these samples also differs substantially. For the ASDEX Upgrade sample, the average material removal rate was about 0.5 μm per single laser pulse. However, for pure graphite sample, this measure was close to 70 nm. This is in agreement with the fact that target reflectance determines the amount of laser pulse power absorbed by the material [18]. Additionally, surface defects are of great importance because they contribute to the decrease of the laser intensity threshold required to initiate vaporization of the surface [5].

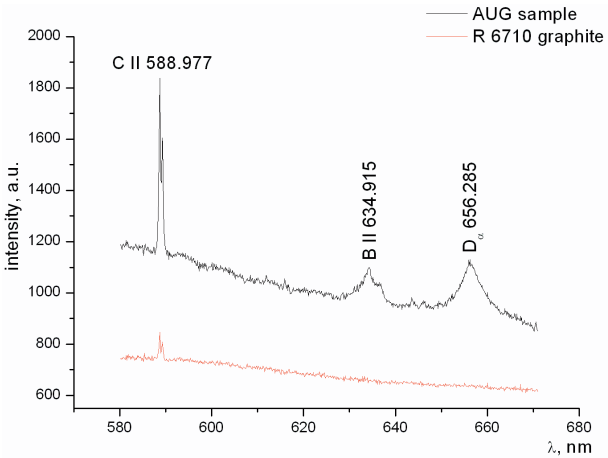

Fig. 3.2. **Comparison spectra of the AUG sample and graphite R7610 sample. One pulse at an energy of 10 mJ was accumulated. D_α 656.285 nm line was not detected in the unexposed sample**

The main element of interest during the investigation of plasma facing materials was hydrogen and/or its isotopes, in the case of ASDEX Upgrade – deuterium line 656.285 nm. D_α 656.285 nm line had been detected in the investigated sample. To be sure that the detected line is exactly deuterium line, spectra of two samples – the ASDEX Upgrade wall sample and graphite R7610 sample which was not exposed to the plasma discharge – were recorded in a narrow spectral window of 620 to 680 nm. The related spectra are represented in Fig. 3.2. The spectra of the unexposed graphite R7610 sample exhibit neither B II (634.927 nm) nor D_α

(656.285 nm) line, which means that these impurities are specific to the samples exposed to plasma discharges.

To follow the evolution of the appearance of deuterium 656.285 nm line in the spectrum, signals in the spectral window of 580–680 nm were recorded. Fig. 3.3 represents spectra corresponding to the 1st, 2nd, 3rd, 5th, and 10th consecutive laser pulses applied at the same spot of the sample.

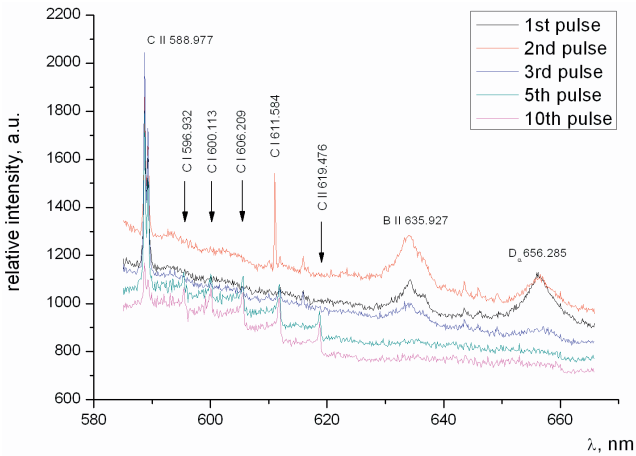

Fig. 3.3. **Appearance of D_α 656.285 nm line [10] in the laser ablation spectrum of the ASDEX Upgrade sample. The numbers in the legend correspond to the numbers of the pulse in the sequence**

The depth profiles of the impurities are represented in Fig. 3.4.

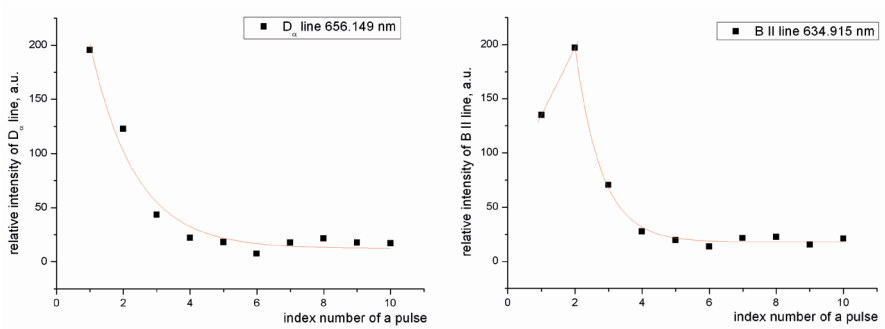

Fig. 3.4. **The depth profile of D_α and B II lines [11] in the laser ablation spectrum of the ASDEX Upgrade sample. The average material removal rate was about 0.5 μm per laser pulse**

Fig. 3.4 demonstrates the spectra obtained after ablation of the AUG sample No. VI. The concentration of deuterium measured with the RBS (Rutherford

Backscattering Spectroscopy) method is 5.36×10^{17} atoms/cm^3, the concentration of boron – 9.78×10^{18} atoms/cm^3. If the ablation removal rate is about 0.5 μm per laser pulse, deuterium can be situated in a depth of 0.5–1 μm inside the bulk material. The concentration of boron, on the other hand, reaches its maximum at about 1 μm depth in the material, then the concentration decreases.

With an increasing number of laser pulses applied to the same spot, the signal of the impurities dropped from high values to the background level, but the signal of the substrate (carbon) remained consistent with the growing number of applied pulses. In atmospheric pressure, the plasma is constrained to small volumes close to the target surface. As the forces sustaining the high pressure region of the plume dissipate, the propagation decelerates and the front edge relaxes. This leads to a portion of plume material being deposited back on the target surface. More volatile elements will condense out of the cooling plasma later than more refractory phases. This condensed material presents a source of the refractory element enriched material which is sampled at the later stages of ablation [19].

3.2 Ablation in Vacuum

The plume characteristics were analyzed by the method of laser-induced spectroscopy. The plume originated from the ablation spot is viewed perpendicularly to the plume propagation direction. The plume parameters depend on the laser fluence, and the observed optical emission varies with distance from the target surface.

3.2.1 Ablation at Different Pressure Values

In order to find the optimal regime for the depth profiling of the plasma facing materials, the experiments were carried out at three different pressures in a vacuum chamber, and at atmospheric pressure. A sequence of 10 laser pulses at a pulse energy of 25 mJ was applied on the targets. The obtained spectra are demonstrated in Fig. 3.5. The legend on each series of laser-induced ablation spectra indicates the pressure value. At each pressure, 10 spectra were recorded, and 3 spectra were registered at atmospheric pressure [20].

The spectra recorded at the pressure value of 10^{-5} Torr are the richest in lines. As the pressure increases, the number of detected spectral lines diminishes. The spectra recorded at atmospheric pressure show the least amount of the spectral lines, but their relative intensities are higher compared to those recorded at lower pressures. Fig. 3.6 illustrates this fact.

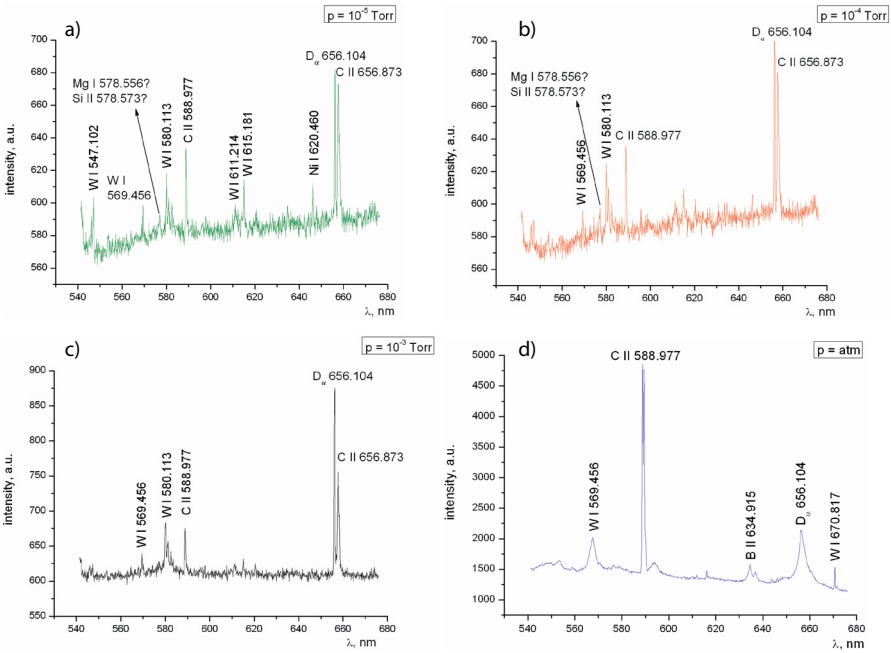

Fig. 3.5. **Spectra of samples taken from ASDEX Upgrade wall material [11]: a) p = 10^{-5} Torr, b) p = 10^{-4} Torr, c) p = 10^{-3} Torr, d) atmospheric pressure. To record each spectrum, the brightest spot of the plume was focused into the optical fibre.**

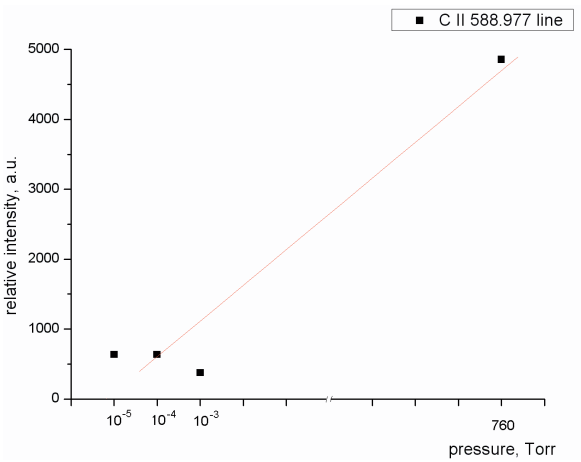

Fig. 3.6. **Intensity of the C II 588.977 spectral line versus pressure in the chamber at the time of recording. The chosen line exhibits the maximum intensity when recorded at the atmospheric pressure**

This observation can be explained with the models of propagation of laser-induced plasma plume demonstrating the development of a high pressure region at the leading edge of the plume [21]. At atmospheric pressure, laser-induced plasma is confined to the region next to the surface of the ablated material. The front edge of the ablation plume interacts with the background gas species, resulting in elevated brightness of the plume in the imaging point.

The spectra recorded at atmospheric pressure show broader spectral lines than those acquired at lower pressure values. When a large number of atoms are concentrated in a small volume to produce a high density of atoms, their interactions with each other cause significant broadening of the width of the emission line in addition to the effect they have upon the decay time of the level causing natural broadening of a spectral line [22]. This occurs when other atoms, ions, or free electrons collide with the excited atom, and it drops down from the excited level before that electron has the opportunity for spontaneous decay.

Fig. 3.5 demonstrates the spectra obtained after ablation of the AUG sample No. IX. The concentration of deuterium measured with RBS is 5.36×10^{17} atoms/cm^3, the concentration of boron – 9.78×10^{18} atoms/cm^3. As in the case of previous experiments, the spectra of ASDEX Upgrade ablation targets show a substantial number of impurities. Apart from tungsten and deuterium lines, Ni I (620.460 nm) is detected at pressure value of 10^{-5} Torr. The spectral line of this element might come from the stainless steel components which are not shielded from the plasma [15]. As for the impurity found at 10^{-4} Torr, both elements might appear in a spectrum. The origin of Si II (578.573 nm) line might be the material of isolation cables. Mg I (578.556 nm) can also be found among the ASDEX Upgrade impurity species [15]. The reason of appearance of B II (634.915 nm) line was explained in the previous section of the paper.

The ablation rate in the described conditions of experiment was approximately the same and reached about 50 μm per pulse. The influence of the pressure is not significant in this case. The spectrum demonstrates the sum of the intensities of the spectral lines, since the experiment was conducted to find the optimal conditions for the ablation procedure. This also explains the reason why D_α line (656.104 nm) is still present in the spectra.

3.2.2 Registration of the Emission Spectra at Various Distances from the Target

Selective detection of the plasma light might give the possibility to enhance detection capabilities; hence, the strong background continuum occurring at early stages of plasma formation can be successfully reduced even without time-resolved detection [23].

Spectra of laser-induced plasma were recorded focusing fibre-optic bundle directly on the ablation spark, then moving the fibre holder along the plasma plume by steps of 0.1 mm (Fig. 3.7). For each step, 10 laser pulses at pulse energy of 20 mJ were accumulated.

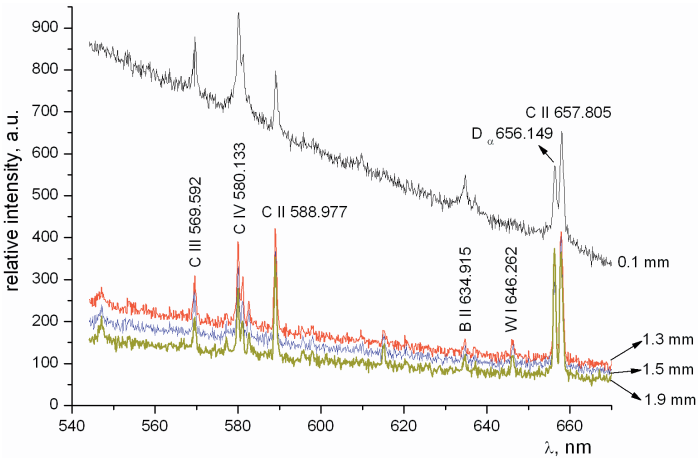

Fig. 3.7. **Spectra of laser-induced plasma at various distances from the ablation spark. For each spectrum, 10 laser pulses at an energy of 20 mJ were accumulated**

The tendency that the background light decreases as the distance from the plasma spark increases is clearly observed. At a distance of about 1 mm from the spark, spectra exhibit similar signal-to-background ratio and spectral contents. As the optical fibre moves away from the ablated surface, the background continuum is significantly reduced compared to the spectra obtained when the brightest point of the plume is focused onto the optical fibre. The signal-to-background ratio is higher in the case when the spectra are recorded in the periphery of the ablation plume. The spectra resemble very much a typical time-resolved LIBS spectrum with low background continuum intensity [24].

Fig. 3.8 demonstrates the dependency of the intensity of CII spectral line (657.805 nm) on the distance of the point where the spectra were accumulated. The relative intensity increases and reaches its maximum at 0.3 mm from the target. Then the decrease of intensity is observed. This can be explained with the fact that the plasma temperature and electron densities decrease when the distance from the ablated sample increases [25].

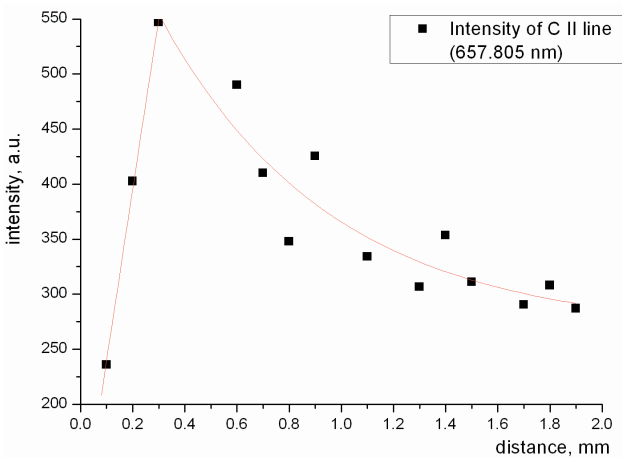

Fig. 3.8. **The relative intensity of the CII spectral line (657.805 nm) at various distances from the ablated sample. The conditions of data accumulation are the same as for the spectra in Fig. 3.7**

Although the plasma temperature might be the highest at the distance of 0.3 mm (see Fig. 3.8) from the target, the optimal distance for the acquisition of plasma spectrum is 1.9 ± 0.1 mm from the target, since signal-to-noise ration is maximal in this case.

As one of the potential applications, accumulation of a certain number of laser shots can be used for tritium desorption from plasma facing materials. To follow the behaviour of spectral lines in spectra, different numbers of laser pulses from 1 to 60 were applied on the surface of the sample (Fig. 3.9 a, b). The spectra were recorded at the distance of 1.9 mm from the ablated target. The obtained spectra show the increase of the relative intensities of carbon lines. In contrast, the signals of impurities – deuterium and boron – decrease with the growing number of pulses. The B II 634.915 nm line is still present in the spectra obtained after 25 consecutive laser pulses, but the signal is equal to the background level in the spectra after 50 laser pulses.

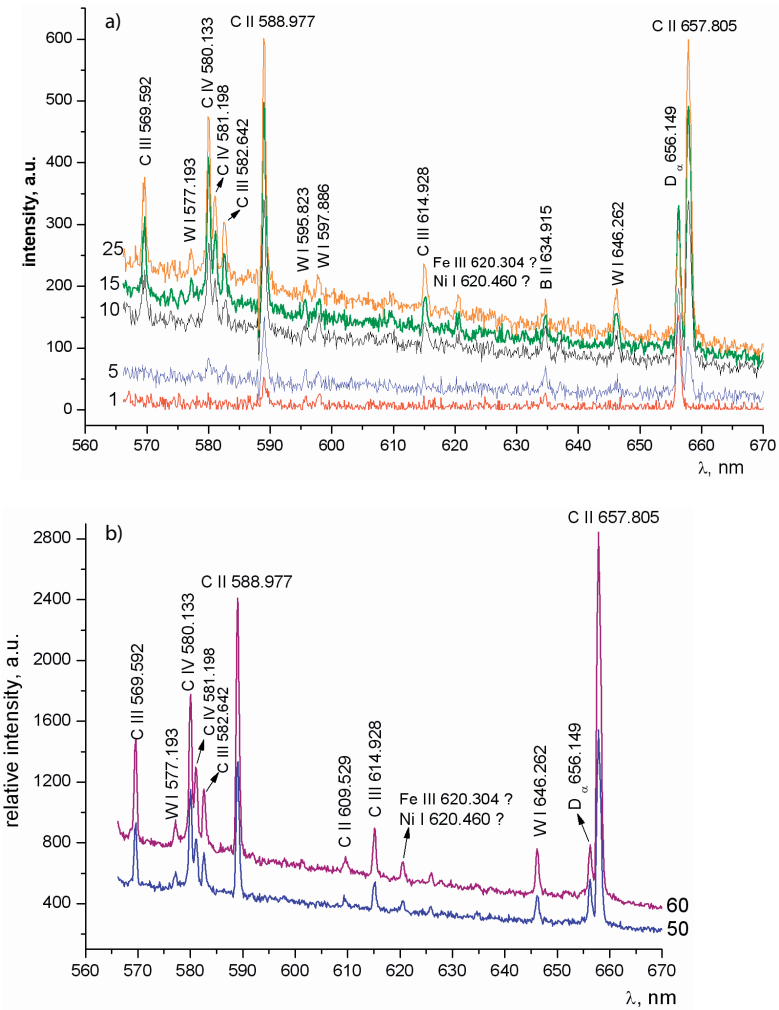

Fig. 3.9 a, b. **The spectra of the ASDEX Upgrade sample after application of a number of laser pulses to the target. The numbers of the spectra correspond to the number of the applied pulses**

Together with laser ablation measurements, ion energy analysis can be performed [26]. The optimal laser treatment regime can be found, so that the spectral information of other impurities can be achieved simultaneously with the process of detritiation.

3.2.3 Ablation at Different Pulse Energies. Depth Profiles

To record the spectral information and the depth profiling of the impurities in the ASDEX Upgrade samples – deuterium and boron – a certain amount of consecutive laser pulses with different pulse energies were applied at the same spot of the target. The spectra were recorded after each applied pulse. As a result, each set of spectra for each laser pulse energy comprises as many spectra as the number of applied pulses. All the measurements were performed at the pressure of 10^{-5} Torr. The spectra are represented in Fig. 3.10.

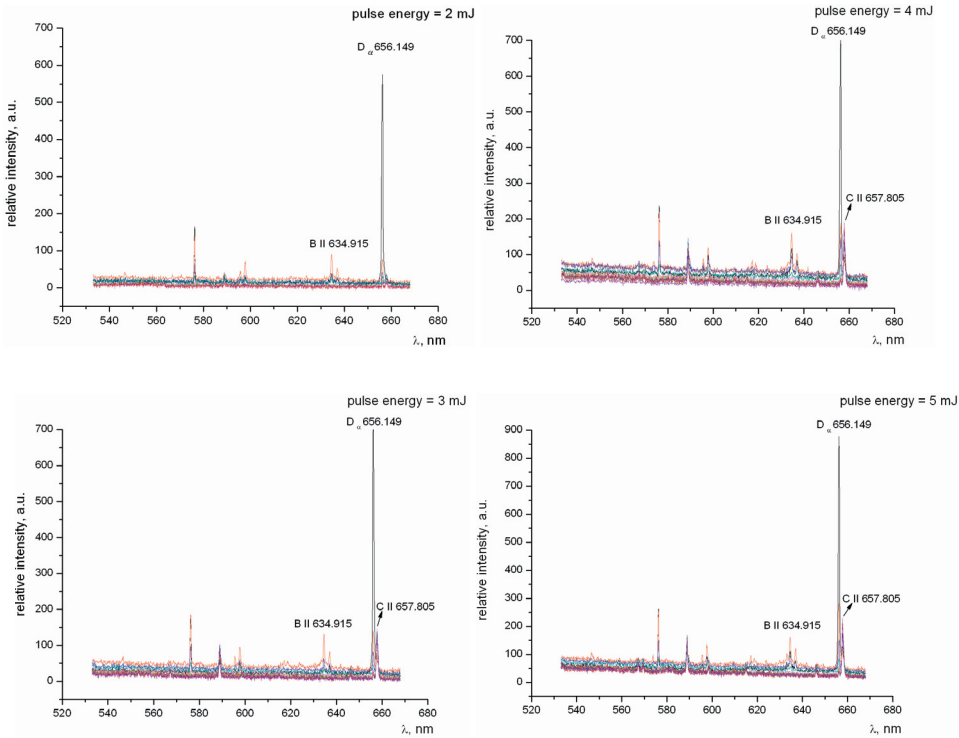

Fig. 3.10. **The spectra of AUG divertor material samples. 9 (in the case of 2mJ pulse) to 14 (in all other cases) consecutive laser pulses were applied at the same spot of the sample. The pulse energy is indicated on each graph. Measurements were performed at the pressure of 10^{-5} Torr**

The events that lead to optical breakdown begin with the formation of a large number of charge carriers in a volume confocal with the laser at or near the surface. When carrier oscillation energy becomes greater than the band gap, bonds can be broken and carriers are multiplied further by avalanche ionization.

Carrier oscillation energy is $E_{osc} = \left\langle \frac{e^2 E^2}{2 m_e \omega^2_{laser}} \right\rangle$, or, in terms of laser wavelength and intensity, $E_{osc} = 9{,}3 \times 10^{-14} I\lambda^2$, where E_{osc} is in eV, laser intensity I is in W/cm^2, and laser wavelength λ is in μm [27]. To ionize a hydrogen atom, the pulse intensity should be of the order of 10^{14} W/cm^2 for the pulse length of the laser used in investigations. The intensities used in the experiment, laser intensity reached values of the order of 10^{13} W/cm^2, which still seems to be sufficient for hydrogen spectral line to appear in the spectra. During the expansion of the plasma plume, it relaxes thermally, and the electron system of hydrogen atom excites by the collisions with plasma particles.

The depth profiles of the impurities are represented in Fig. 3.11.

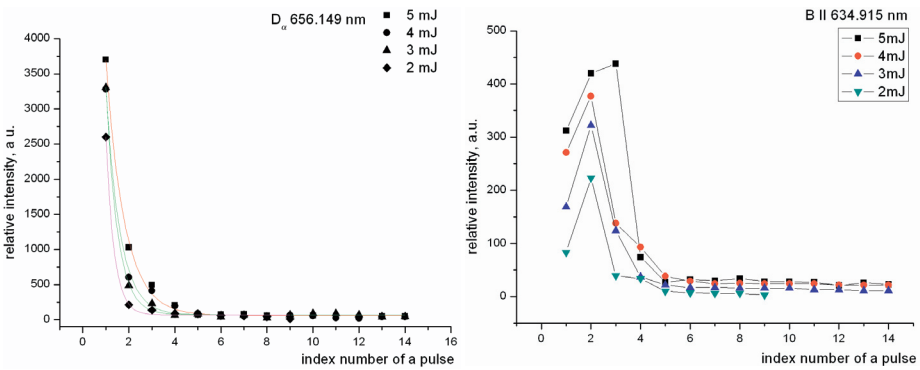

Fig. 3.11. **Depth profiles of D_α and B II lines at different energies of laser pulses. The pulses were applied one by one at the same spot of the sample, and the spectra were recorded after each pulse. The index number of a pulse in the sequence is shown on the X axis**

Fig. 3.12 shows the behaviour of the C II line (λ = 657.805 nm) at different laser fluences.

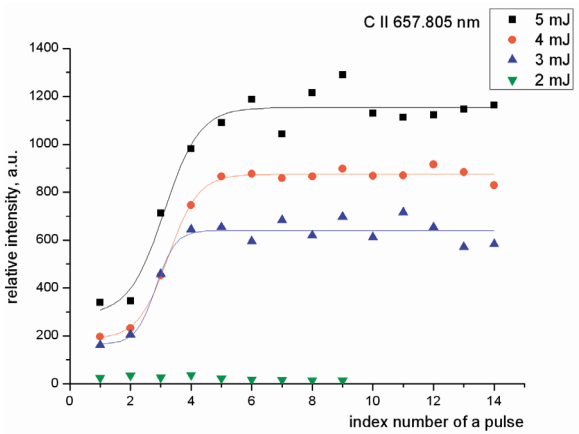

Fig. 3.12. **Evolution of the C II line (λ = 657.805 nm) with the number of applied pulses at different pulse energies. The pulses were applied one by one at the same spot of the sample, and the spectra were recorded after each pulse. The index number of a pulse in the sequence is shown on the X axis.**

As graphite is the basic material of the ASDEX Upgrade samples, the intensity of the C II 657.805 nm line is growing with the number of applied pulses, and after the 5th pulse the signal remains approximately constant. The intensities corresponding to the ablation at a pulse energy of 2 mJ are nearly constant and equal to the background level, as the ablation threshold of the graphite exceeded very little.

To investigate the shape of the ablation craters, as well as the ablation rate in the case of each laser pulse energy corresponding to those used in this experiment, 100 pulses were applied on the surface of the ASDEX Upgrade tile and then measured with the profilometer. The ablation pits of the relevant pulse energies are represented in Fig. 3.13.

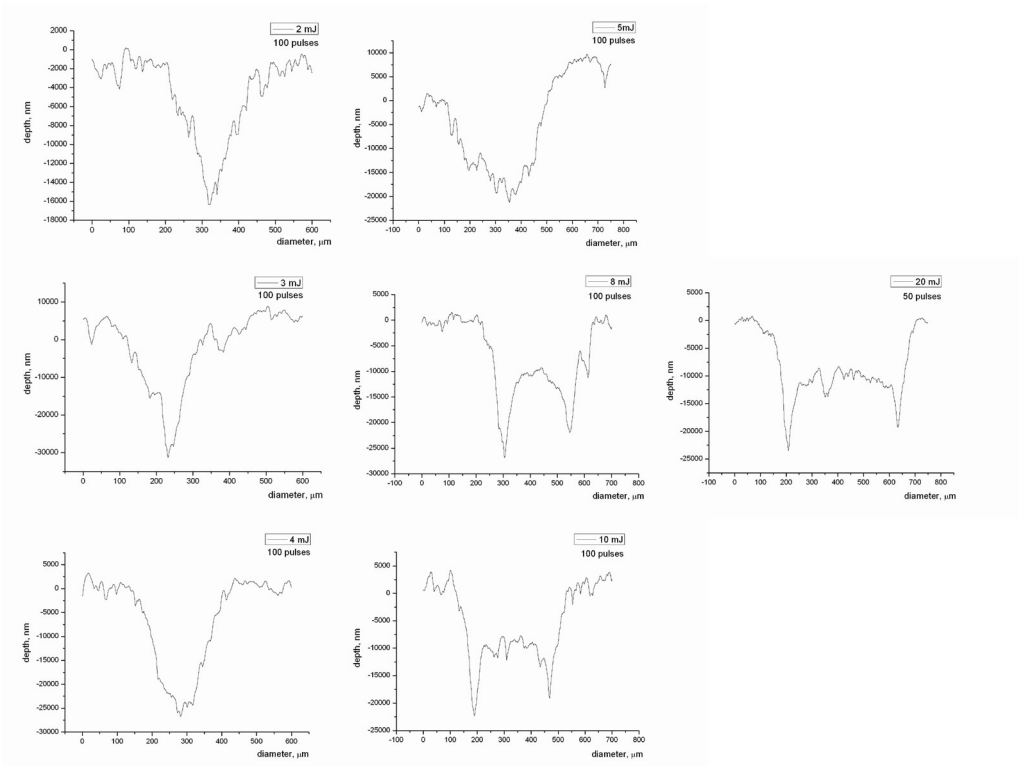

Fig. 3.13. **Profiles of the ablated craters. Pulse energies and accumulated shots are indicated on each graph**

At higher intensities of the incident laser beam, one can notice a specific track around the edge of the ablation crater (see Fig. 3.13 for 8, 10, and 20 mJ pulses). We believe that this is the result of the edge diffraction occurring during the ablation process [28].

During laser ablation, the topography of the sample surface that is treated by laser changes. The edges of the laser ablation pits as well as the untreated surface of the sample are smoother in comparison with the interior of the hole. Hence, laser ablation creates regions with different absorption coefficients, and they are higher at the rougher spots of the surface, forming a kind of mask. This mask works as a diffraction pattern, creating a profile corresponding to the intensity distribution occurring as a result of straight edge diffraction.

Figures 3.10–3.12 demonstrate the spectra obtained after ablation of the AUG sample No. X. The concentration of deuterium measured with RBS is

5.84 × 10^{17} atoms/cm^3, the concentration of boron – 1.11 × 10^{19} atoms/cm^3. Considering the ablation removal rate in the case of applying 2–5 mJ, the results correspond to those obtained during the measurements at atmospheric pressure. Almost all the deuterium found in the ASDEX upgrade samples is released from the depth of 0.5–1 μm inside the bulk material. The concentration of boron is maximal at 1 μm depth in the material, then the concentration decreases.

The depth of the craters does not exhibit considerable changes with the growth of the pulse energy, although the diameter changes significantly.

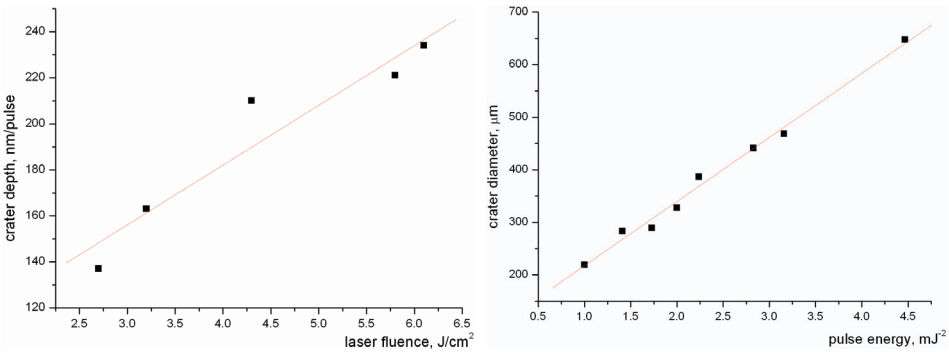

Fig. 3.14 a, b. **The amount of ablated matter changes with the pulse energy of a laser beam. Crater depth (a) is plotted as a function of laser fluence, and crater diameter (b) as a function of the square root of the pulse energy**

Similarly to the fluence dependence on the crater depth, the discontinuity in the crater volume data is observed in the low fluence regime, although it is not so pronounced. The discontinuity in the fluence dependence of the crater depth and crater volume; hence, the onset of significant material removal was associated with the ablation threshold.

The growth of the diameter of the ablation crater scales as a square root of the pulse energy. The reason is most likely the mass transport. When fluence increases, a significantly larger volume of material melts. Tight focusing leads to steep temperature gradients in the lateral dimensions, and due to the possibility of hydrodynamic flow [29], the final surface morphology is not expected to be an accurate representation of the local fluence.

Besides the shape of the ablation craters, another reason why the depth profiling can be slightly imprecise is the surface of the sample itself. Fig. 3.15 shows the intact surface of the ASDEX Upgrade sample scanned with profilometer.

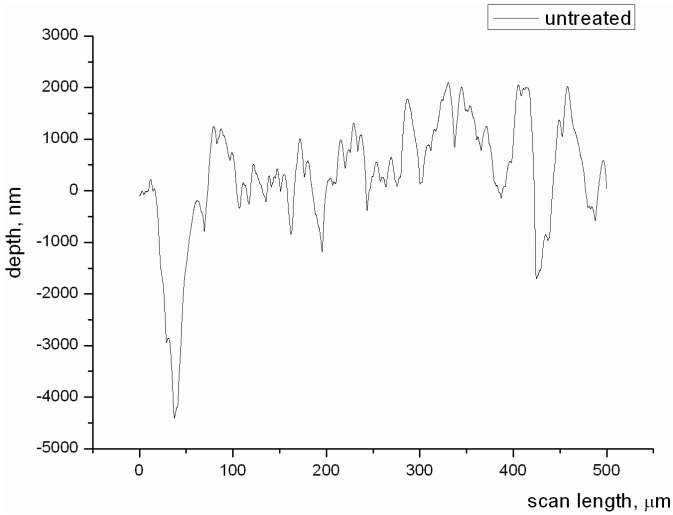

Fig. 3.15. **The surface of the AUG sample untreated with laser ablation**

The sample was not treated with laser. The scan shows relatively deep pores which can hinder an accurate analysis of the depth of the ablation craters. The erosion of the plasma facing materials inside the tokamak contributes a lot to the modification of their surface, but the specification of the R6710 graphite shows that the material is relatively porous, and individual pores can reach 0.6 μm.

Theses

1. Two ablation mechanisms have been observed. Before a certain laser fluence threshold is reached (4.3 J/cm^2 with pulse length 0.1 ns), ablation processes are determined by melting of the material. When this threshold is exceeded, ablation processes are determined by ionization of atoms in the electric field of the laser pulse.

2. Different mechanisms of the ablation processes are expressed in the specific shapes of the ablation craters. When a certain laser fluence threshold (4.3 J/cm^2) is exceeded, a particular shape of the ablation crater features laser beam diffraction on the crater wall described by straight edge diffraction. At laser fluences above the threshold, the ablation crater is bell-shaped.

3. There is an optimal distance from the surface of the ablated material at which the intensity of a spectral lines of certain impurities is maximal with respect to the background continuum light. It is determined by the thermal relaxation of

plasma as it expands, as well as by the excitation of the electron system of the impurity atom (hydrogen) during its collisions with plasma particles.

Summary of the Results

The main results obtained in this investigation are the following:

- experimental equipment for laser-induced breakdown spectroscopy analysis of plasma facing materials has been developed at the Optical Spectroscopy Laboratory at the Institute of Solid State Physics;
- a methodology for laser ablation of impurity depth profiling has been developed;
- the accumulation depth of the impurities has been estimated.

The optimal pressure for laser ablation measurements was determined to be 10^{-5} Torr. The spectra recorded at the pressure value of 10^{-5} Torr are the richest in lines. As the pressure rises, the number of detected impurities declines. The spectra recorded at atmospheric pressure show the smallest number of spectral lines, but their relative intensities are higher compared to those recorded at lower pressures. The spectrum recorded at 10^{-5} Torr is well-resolved and exhibits the largest number of lines among the pressure values used in the investigations. The spectral lines of the impurities of interest are not subjected to such pronounced broadening as in the case of the experiments at atmospheric pressure.

The tendency that the background light decreases when the distance from the plasma spark increases is clearly observed. At a distance of about 1 mm from the spark, spectra exhibit similar signal-to-background ratios and spectral contents. As the optical fibre moves away from the ablated surface, the background continuum is significantly reduced compared to the spectra obtained when the brightest point of the plume is focused onto the optical fibre. The signal-to-background ratio is higher in the case when spectra are recorded in the periphery of the ablation plume. The optimal distance from the ablated target for the recording of spectral information is deduced to be 1.9 ± 0.1 mm. This distance is determined by the thermal relaxation of plasma as it expands, as well as by the excitation of the electron system of the impurity atom (hydrogen) during its collisions with plasma particles.

The investigations of the depth profiles of the ASDEX Upgrade samples were performed at different pulse energies, and two ablation mechanisms were observed. When a certain laser fluence threshold (4.3 J/cm^2) is exceeded, a particular shape of the ablation crater features laser beam diffraction on the crater

wall described by straight edge diffraction. At laser fluences above the threshold, the ablation crater is bell-shaped.

The mechanism of laser ablation of the divertor material of the ASDEX Upgrade tokamak can be described as follows: when the laser pulse interacts with the target surface, the elements of the impurities appear in the spectrum. This happens as the impurity layer on the ASDEX Upgrade target is removed in the ablation process. The layer can be as thick as (3 ÷ 4) × material removal rate, which is about 0.5 μm per laser pulse (Fig. 3.16).

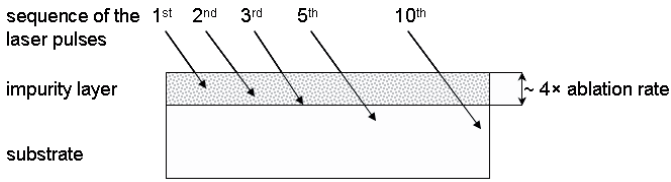

Fig. 3.16. **The process of the ablation of impurities in the ASDEX Upgrade target**

Almost all hydrogen was released from the ASDEX Upgrade tiles at the depth of 0.5–1 μm in the process of laser ablation. The concentration of boron reached its maximal value at the depth of 1 μm and decreased with the increase of the depth of the ablation crater.

Practical Significance

The results of the thesis research are of practical significance in the following aspects.

1. The method of laser-induced ablation analysis of the impurities in plasma facing components allows to detect any chemical elements; therefore, it can be used for detection of hydrogen isotopes in plasma facing components.

2. Laser-induced ablation spectroscopy can be performed as remote analysis. This is essential for plasma facing materials because detection of hydrogen isotopes is one of the crucial tasks for the analysis of the surface of plasma facing components, and because the irregular shapes of components form spaces which are difficult to reach.

3. The results show that laser-induced ablation spectroscopy can also be used for detritiation of plasma facing components.

Conclusions (Future Applications)

1. The results of this work stress the importance of developing a fast, relatively cheap (in comparison with the existing techniques) method for surveying plasma facing components directly in the chamber of thermonuclear fusion reactor.

2. Together with laser ablation measurements, ion energy analysis can be performed. Measuring ion signals before and after laser ablation, a minimal number of laser pulses can be determined to remove deuterium or tritium from the co-deposited layer completely. An optimal laser treatment regime can be found for spectral information on other impurities to be obtained simultaneously with the process of detritiation.

3. Determination of concentration of the impurities in plasma facing materials requires concentration calibration standards. Since the materials needed for such standards are too specific, they are not commercially available. Therefore, a cross-calibration which can provide both qualitative and quantitative depth profiling of the impurities can be useful.

4. It was proven that deuterium feature is almost equal to the background level after the 4th laser pulse is applied to the sample. This can be improved without increasing the intensity of the laser beam applied to the target. A dual-pulse laser ablation can be used. Improvement of the limit of detection under double-pulse excitation showed nearly an order of magnitude compared to what can be achieved with single-pulse ablation.

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List of Author's Scientific Publications

1. J. Butikova, A. Sarakovskis, B. Polyakov, I. Tale. Laser ablation for analysis of nanoscale layers. *Journal of Physics: Conference Series*, Vol. 93, 2007, p. 012043.
2. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced breakdown spectroscopy for determining impurity content and depth profile in plasma facing materials. Submitted for publication in *Optics and Lasers in Engineering.*
3. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced ablation spectroscopy for deuterium detection in plasma facing components. Submitted for publication in *Spectroscopy Letters.*
4. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced plasma spectroscopy of plasma facing materials. 35th EPS Conference on Plasma Physics, Hersonissos, Greece, 9–13 June 2008, ECA, Vol. 32, P-2.011.
5. B. Polyakov, A. Petruhins, J. Butikova, A. Kuzmin, I. Tale. Some aspects of pulsed laser deposition of Si nanocrystalline films. *Eur. Phys. J. Appl. Phys.* 48, 2009, p. 20502.

Participation in Conferences

1. J. Butikova, I. Tale. Laser Ablation Spectroscopy for Concentration of Impurity Elements. ISSP 21st Scientific Conference, Abstract Book, 2005, Rīga, Latvia.
2. J. Butikova, I. Tale. Laser Ablation Spectroscopy for Impurity Depth Profiling in Plasma. Book of Abstracts of the 7th International Summer School-Conference "Advanced Materials and Technologies". Aug 2005, Palanga, Lithuania, p. 129. ISBN 9955-09-894-5
3. J. Butikova. Laser Ablation Spectroscopy for Impurity Depth Profiling and Concentration Imaging in Plasma. Internal seminar at the Department of Material Sciences, Max Planck Institute of Plasma Physics, October 2005, Garching, Germany.
4. J. Butikova. Rutherford backscattering measurements for laser induced breakdown spectroscopy standards. ISSP 22nd Scientific Conference, Abstract Book, 2006, Rīga, Latvia, p. 41.
5. J. Butikova. Laser Induced Breakdown Spectroscopy. Internal seminar at the Department of Material Sciences, Max Planck Institute of Plasma Physics, October 2006, Garching, Germany.
6. J. Butikova, A. Sharakovsky, I. Tāle. Development of Equipment for Laser Ablation Spectroscopy. ISSP 23rd Scientific Conference, Abstract Book, 2007, p. 53, Rīga, Latvia.
7. J. Butikova, A. Sarakovskis, I. Tale. Laser Ablation Spectroscopy of Plasma-Facing Materials. Development in Optics and Communications 2007, Abstract Book, p. 15, Rīga, Latvia.
8. J. Butikova, A. Sarakovskis, B. Polyakov, I. Tale. Laser ablation for analysis of nanoscale layers. International Baltic Sea Region Conference "Functional Materials and Nanotechnologies", April 2–4, 2007, Rīga, Latvia.
9. J. Butikova, A. Sarakovskis. Laser ablation spectroscopy for impurity depth profiling and concentration imaging in plasma. EFDA Workshop, October 2007, Rīga, Latvia.
10. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced breakdown spectroscopy application for determining impurity content and depth profile in the plasma facing materials. Conference Proceedings of the 13th International Conference on Fusion Reactor Materials, December 10–14, 2007, Nice, France.
11. J. Butikova, A. Sarakovskis, I. Tale. Laser ablation spectroscopy of the first wall materials of *ASDEX Upgrade* tokamak. ISSP 24th Scientific Conference, February 2008, Rīga, Latvia.
12. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced ablation spectroscopy for deuterium detection in plasma facing components. International Baltic Sea Region Conference "Functional Materials and Nanotechnologies", April 1–4, 2008, Rīga, Latvia.
13. J. Butikova. Laser-induced breakdown spectroscopy of plasma facing materials. Invited lecture at the Faculty of Physics, Vilnius University, May 28th, 2008.
14. J. Butikova, A. Sarakovskis, I. Tale. Laser-induced ablation spectroscopy of plasma facing materials. 35th EPS Conference on Plasma Physics, June 9–13, 2008, Hersonissos, Greece.

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