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Study of the feasibility of applying laser-induced breakdown spectroscopy for in-situ characterization of deposited layers in fusion devices


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Abstract
This paper presents a feasibility study of laser-induced breakdown spectroscopy (LIBS) for the development of an in-situ diagnostic for the characterization of deposition layers on plasma-facing components in fusion devices. Preferentially, LIBS would be applied in the presence of a toroidal magnetic field and under high vacuum conditions. The impact of the laser-energy densities on the laser-induced plasma parameters and correspondingly on the number of emitted photons and on the reproducibility of the LIBS method has been studied in laboratory experiments and in TEXTOR on fine-grain graphite (EK98) as well as on bulk W samples coated with carbon and metallic-containing deposits. The effect of magnetic fields and of ambient pressures in the range from $2 \times 10^{-4}$ Pa to 10 Pa on the carbon plasma plume produced by the LIBS technique has been studied on TEXTOR between plasma pulses. The possibility of applying this method to ITER is discussed.

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

1. Introduction

Tritium retention inside the vacuum vessel has emerged as a potentially serious constraint in the operation of ITER [1]. ITER is currently designed with a beryllium-clad first wall, tungsten (W) on the divertor upper baffle and dome and two options for the high-power divertor areas: carbon fibre composites to begin with, to be changed most probably to a full W divertor in the D–T operation phase [2]. The tritium inventory with such choices of facing materials in ITER is expected to be dominated by co-deposition with carbon and beryllium, with a minor contribution from T retention in the bulk of the W tiles [3]. Knowledge of the distribution, thickness and composition of these layers, which will be strongly inhomogeneous, especially in the poloidal direction, is essential for the safe operation of ITER. In-situ characterization of deposited layers (tritium quantity and surface distribution, thickness and composition) is of major importance for the operation of fusion devices with respect to safety [4].

A large majority of the data on fuel retention in present devices is based on post-mortem analysis of tiles that are taken out of the device after a certain time of operation, thus averaging over a variety of wall and plasma conditions. These measurements become more and more difficult with the use of actively cooled tiles, in particular in the activated phase when tritium is used together with Be tiles and neutron activation occurs. For safety reasons, only 700 g of tritium is permitted inside the ITER vessel, which requires the monitoring of tritium practically between each plasma discharge to take a decision on the permission to initiate the next plasma pulse. Laser-based methods combined with optical spectroscopy during discharges and laser-induced breakdown spectroscopy (LIBS) between discharges are commonly proposed [5, 6].
to provide in-situ data on the amount of material deposition and on fuel retention. The tile removal is unnecessary and the method is compatible with tritium, beryllium and neutron activation. Moreover, it can also survey the effect of cleaning methods to allow a safe start of the next plasma discharge.

In this paper, we will restrict the discussion on laser-based methods to LIBS. The LIBS method is a well-known laser method for the quantitative analysis of surface matter composition [7] and is used already as a routine method for several practical applications. However, almost all LIBS applications have been performed at atmospheric pressure or in some residual gaseous environment. For fusion applications, the method would benefit if it could operate under vacuum conditions for use between plasma pulses. The basic idea of the LIBS method for in-situ characterization of first-wall surfaces in fusion devices is to heat and evaporate material at different spots on the first wall by intensive laser radiation between discharges. Laser radiation focused on the target surface results in ablation of the surface material, which leads to the formation of a laser plasma if the laser intensity and the material release exceed a critical level. LIBS is proposed as a candidate diagnostic for the characterization of the wall conditions in ITER [5, 6] and has recently been integrated into the baseline design. In the current ITER design, the LIBS diagnostic will be performed during in-vessel inspections and will be mounted on a remotely operated arm. However, such an implementation is restricted by many technical requirements of the remote arm, taking into account the constraints of such a multipurpose remote system as well. Alternatively, we propose the application of LIBS from a port-plug-based laser, which should be operated under vacuum. This will provide an in-situ method that can be applied routinely between shots, while the superconducting coils still produce a permanent toroidal magnetic field. Under such special conditions in fusion devices, the following issues should be addressed:

- the applicability of the LIBS method under high vacuum conditions,
- the influence of different material compositions and properties on the LIBS plasma parameters,
- the reproducibility/stability and sensitivity of LIBS signals,
- the influence of the ambient pressure on the LIBS plasma parameters, and
- the influence of permanent magnetic fields on the LIBS signals.

2. Experimental setup

The feasibility of the LIBS method has been studied at first in laboratory experiments in a high vacuum chamber with a base pressure below $10^{-4}$ Pa. The laser-induced plasma plume from a Q-switched ruby laser with 1 J maximum energy and 15 ns pulse duration is observed in a direction parallel to the target surface within a volume of about 1 cm$^3$ and is analysed by a high-resolution cross-dispersion Echelle spectrometer (resolving power $\lambda/\Delta\lambda \approx 20 000$) [8] in the spectral range between 375 and 715 nm. To avoid the cumulative effects of the repeated pulses (e.g. heating up of the surfaces), measurements were carried out at a laser repetition rate below 0.2 Hz.

In addition, to study the influence of the background pressure and of a permanent magnetic field on the LIBS plasma parameters, LIBS measurements were started in a second experiment, on the TEXTOR tokamak during a recent experimental campaign. A Q-switched Nd:YAG laser at a fundamental wavelength of 1064 nm with a peak energy of 2.5 J and pulse duration of about 7 ns has been used. The laser can also be operated in the second and third harmonics. To simulate the ITER-like conditions, a full mirror-based beam line of about 20 m optical path has been installed, which directs the laser light onto the samples and limiter objects positioned in the TEXTOR torus through the limiter lock system [9]. In the present paper, samples made of a fine-grain graphite EK98 were used. A simple plano-convex quartz lens focused the laser beam onto a spot of about 4 mm diameter at the sample surface. The laser-induced plasma plume is recorded in a tangential view from the side by a two-dimensional (2D) camera and the high-resolution cross-dispersion Echelle spectrometer mentioned above.

3. Result and discussion

3.1. General characterization of the LIBS plasmas

To study the feasibility of the LIBS method, fusion relevant mixed layers have been deposited on the top of bulk W samples by various techniques such as glow discharge plasma deposition and magnetron sputtering. These layers have been ablated and investigated by LIBS in laboratory experiments and in the TEXTOR vessel. Figure 1(a) shows spectroscopic LIBS signals of C, H, Cr and W emission lines of different ionization stages from a 2.5 μm thick a-C:H layer on W with a Cr interlayer obtained in the laboratory experiment. The carbon deposit is removed within about four laser shots when the interlayer is reached, identified by the appearance of the Cr and W signals. This corresponds to the removal rate of $\approx 0.6 \text{μm pulse}^{-1}$ and is in agreement with material ablation rates observed in earlier experiments, which will be discussed later in the paper, for laser fluences larger than 8 J cm$^{-2}$. As an example, the spectra of the laser-induced plasma in front of a W sample coated with an a-C:H layer at an energy density of 18 J cm$^{-2}$ are shown in figure 1(b). In this case, LIBS was applied in a laboratory experiment. Lines of singly and doubly ionized carbon atoms as well of neutral Cr atoms can be identified. The electron density of the LIBS plasma has been estimated by measuring the Stark broadening of the spectral lines of C$^+$ ions (see figure 1(c)). The density, averaged over the entire lifetime of the laser plasma plume, varies between $5 \times 10^{21}$ and $6 \times 10^{21}$ m$^{-3}$ at an energy density of about 18 J cm$^{-2}$.

3.2. Impact of the laser-energy densities on the laser-induced plasma parameters

The investigation of the impact of the laser-energy densities on the laser-induced plasma parameters and correspondingly on the number of emitted photons and on reproducibility is one of the main topics of the feasibility study of the LIBS method for ITER application. In figure 2(a), the ablated carbon mass normalized to an area of 1 cm$^2$ shows a linear

Figure 1. (a) Normalized LIBS signals of C, H, Cr and W emission lines of a 2.5 µm thick a-C:H layer on W with a Cr interlayer. (b) LIBS spectrum at a laser-energy density of 18.0 J cm⁻² in the fourth laser pulse at the same irradiation location a-C:H layer on the tungsten. (c) The profiles of Hα and CII emission lines used for electron density measurement by Stark broadening. The red line corresponds to the Voigt fit of the normalized emission lines.

dependence on the energy fluence up to 7 J cm⁻² and saturates at about 10 J cm⁻². A possible reason for the saturation of the craters is the vapour shielding induced by the intense laser ionizing and heating the evaporated cloud. The material ablation rates, obtained from depth profiles of laser-produced craters and from EK98 graphite density of 1.85 g cm⁻³, are in the range ~0.3–0.6 µm pulse⁻¹ at the energy density range \( F \approx 2.6–7.1 \text{ J cm}^{-2} \) and saturate at about ~0.7 µm pulse⁻¹ for higher energies. More information about ablation experiment can be found in [10].

Additionally, it was observed that stable conditions of the laser plasma have been achieved only at laser-energy densities above 8 J cm⁻²; otherwise the fluctuation of a single emission line from pulse to pulse can reach about 100% independently of the type of C-layer. Therefore, the operation at \( F < 8 \text{ J cm}^{-2} \) is affected by uncertainties that are too large for the analysis of the ablated layer in a single laser pulse. Above this value for the energy density, the fluctuation level of e.g. the CII lines is below ±10%, indicating the high reproducibility of the CII signal from pulse to pulse.

![Figure 2](image_url)

Figure 2. (a) Dependence of area-normalized ablated graphite mass on energy density. (b) The conversion factor \( C_f \) (the ratio of the ablated atoms to the number of CII photons) as a function of laser-energy densities.

For the experimental determination of the conversion factor \( C_f \), the ratio of the ablated C atoms \( (N^C) \) to the number of CII line photons \( (I_{\text{photons}}) \) in full solid angle \( (4\pi) \), the LIBS method has been applied to well-known prepared samples, supported by absolute calibrated spectroscopy. Laboratory results show that for \( F \geq 10 \text{ J cm}^{-2} \) on fine-grain graphite (EK98), the conversion factor \( C_f \) resulting from the single ionized C (426.7 nm, 2s²3d–2s²4f) is given as \( C_f = N^C/I_{\text{photons}} \approx 10^6 \). Under absorption of lower laser-energy densities, the plasma parameters \( (n_e \text{ and } T_e \text{ of the laser-induced plasma}) \) and, correspondingly, the species distribution show strong dependence on laser-energy density and this conversion factor \( C_f \) varies significantly as shown in figure 2(b). A nearly constant value has been observed for energy densities above 10 J cm⁻². Thus operation at energy densities \( \geq 10 \text{ J cm}^{-2} \) is preferable to keep the \( C_f \) factor at a constant level and, at the same time, to ensure a fluctuation level of the observed CII lines below ±10%.

The parameters of the LIBS diagnostic for ITER can be estimated from an experimentally determined \( C_f \) factor. With a laser spot size of 1 cm² and a C areal density of \( \approx 5 \times 10^{22} \text{ C m}^{-2} \) released during the LIBS, we would get \( \approx 5 \times 10^{12} \) CII emission photons per laser pulse with energy densities above 10 J cm⁻². The number of photoelectrons \( N_{\text{el}} \) measured with a detector is given by

\[
N_{\text{el}} = \left[ N^C/C_f \right] \times T \frac{\Delta \Omega}{4\pi} \eta,
\]

where \( C_f = 10^6 \) is the conversion factor, \( \Delta \Omega \) is the solid angle of optical system, \( T \) is the transmission factor of the optical system and \( \eta \) is the quantum yield of the detector.

The optical parameters are taken from the proposed experimental arrangement for laser diagnostics on ITER [11]:

3
**3.3. The influence of the background pressure on the LIBS plasma parameters**

The laser-induced plasma parameters may also be influenced by the base pressure in the high vacuum chamber. This fact can be used to increase the sensitivity of the LIBS method by selecting the optimal pressure and type of the gas used. A study of the influence of ambient pressure on the LIBS plasmas in the pressure range from $3 \times 10^{-4}$ to 10 Pa has been performed on TEXTOR. Fine graphite EK98 samples were placed perpendicularly to the direction of the laser beam. The electron density of the laser-induced plasma has been estimated by measuring the Stark broadening of the spectral lines of C$^+$ ions (426.7 nm, 2s$^2$3d–2s$^2$4f). Figure 3 shows the normalized CII emission lines fitted by Voigt profiles for two selected pressures, $2 \times 10^{-4}$ and 10 Pa, without the presence of a magnetic field. The time-averaged density for both cases is about $1.6 \times 10^{21}$ cm$^{-3}$ and does not show any significant difference. Thus, the LIBS application in such conditions does not show any preferable ambient pressure for increasing the sensitivity of the method.

**Figure 3.** Influence of ambient pressure on electron density of LIBS plasma in TEXTOR.

![Figure 3](image)

**3.4. Study of the influence of magnetic fields on the LIBS signals**

The presence of a magnetic field during the expansion of the laser-produced plasma may affect its emission characteristics.
A study of the influence of varying magnetic fields up to 2.5 T on the LIBS plasmas has been performed on TEXTOR at an ambient pressure of about 10^{-4} Pa. The laser beam was focused on a fine-grain graphite EK98 sample located in the limiter lock system. Figure 4 shows the CII emission light (514 nm, 2s2p(3P)^3s–2s2p(3P)^3p) without and with the presence of a magnetic field (B = 2.25 T). The CII emission was selected with an interference filter and recorded by a 2D-CCD camera from a tangential view from the side. The LIBS plasma in a magnetic field of 2.25T shows about 50% enhancement in the CII emission line intensity (see the intensity profile integrated along the sample surface in the lower panel of figure 4). In addition, an increase of the laser-induced plasma plume size from about 6 to 10 mm has been observed. Enhancement of the LIBS signals in aluminum plasma in the presence of a magnetic field (B = 0.64 T) has been observed in [12]. The temperature of the plume was found to increase due to the conversion of the kinetic energy of the plasma plume into Joule heat. Additionally, the electron density nearly doubled in the presence of a field at early times. Thus, first LIBS experiments in the presence of a magnetic field delivered the good message that there is no deterioration of the sensitivity of the LIBS technique (the CII emission does not show any reduction). However, further studies of the application of LIBS are required to investigate the influence of the tokamak magnetic field on the laser-induced plasma parameters. Correspondingly, conversion factors for each detected species should be experimentally determined for the entire range of toroidal magnetic field in fusion devices.

4. Summary and conclusion

The feasibility of the LIBS method proposed for ITER with laser injection from a port plug under vacuum conditions as well as with a magnetic field has been studied in laboratory experiments and on the TEXTOR tokamak between plasma discharges.

To keep the ablation rate at a constant value and to ensure, at the same time, the LIBS signals reproducibility, it is preferable to operate with laser-energy densities of about 10 J cm^{-2} (for the laser system with about 10 ns pulse duration). Assuming a realistic laser optical path transmission of 50% and a laser spot of 0.25 cm², laser systems with a laser energy of 5 J and a pulse duration of 10 ns will fulfil the requirements of an application to ITER. These energy densities will allow us to resolve the LIBS signal with a good photoelectron statistic N_{el}^{-0.5}, when about 10^{18}C atoms (content of C atoms in a 100 nm thick layer) will be ablated from a C layer. As expected, one full power ITER pulse will build up a deposition in the range of 1 μm.

Such thick layers, one order of magnitude above the estimated detection limit of the LIBS, demonstrate the applicability of the proposed LIBS technique in ITER for monitoring carbon deposits. Further investigations to extend the LIBS diagnostic to the monitoring of other wall materials and fuel species are expected in laboratory experiments as well on TEXTOR.

The influence of the ambient pressure on laser-produced carbon plasma properties was investigated on TEXTOR for pressures ranging from 2×10^{-4} to 10 Pa. In this pressure range, the absolute values of CII integrated emission shows only moderate changes (below 15%) and thus do not show any preferable baseline pressure for an increase of the method’s sensitivity. The electron density of the laser-produced plasma, evaluated from the Stark broadening of the spectral lines of C⁺ ions, does not show a significant influence of the ambient pressure in the observed range.

The effect of varying magnetic fields up to 2.5 T on the carbon plasma produced by the LIBS technique has been studied on TEXTOR. Enhancement of the intensity has been observed for spectral lines from singly ionized carbon atoms. In addition, an increase of the laser-induced plasma plume size from about 6 to 10 mm in the presence of a magnetic field was observed.

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