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Detection by LIBS of the deuterium retained in the FTU Toroidal Limiter*

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Abstract

In this paper the Laser Induced Breakdown Spectroscopy (LIBS) measurement of the deuterium (used as a proxy for tritium) retained in and the surface elemental composition of the FTU Mo (TZM) toroidal limiter tiles, carried out from remote (~ 2.5 m) during machine maintenance, are reported. Single pulse technique has been used with the FTU vessel under high vacuum or in Nitrogen or Argon atmosphere. In vacuum experiments D_α and H_α lines have been detected with good resolution, while in Ar atmosphere (5 x10^4 Pa) the two lines were partially overlapped due to Stark broadening. First results of measurements in N_2 atmosphere (10^5 Pa) showed no presence of D_α and H_α lines. These measurements were also carried out for supporting the foreseen use of a robotic arm for an extended LIBS analysis of retained deuterium in the FTU vessel components.

Key words: LIBS, FTU tokamak, toroidal limiter, deuterium retention

1. Introduction

The quantitative detection of tritium retained in the ITER in vessel components is mandatory for deciding if the machine operation must be stopped and the exceeding tritium removed. Laser Induced Breakdown Spectroscopy [1] is a suitable, with micro-destructive characteristics, in situ diagnostic for detecting retained tritium. It is still under discussion whether the LIBS analysis from outside the vacuum chamber is sufficient to extrapolate the measured quantity to the entire ITER wall or a robotic arm equipped with LIBS system should be employed in the vented machine to analyze a more consistent area of the vessel.

LIBS measurements under vacuum were already carried out in FTU to check the dependence of the emitted line intensities on the applied toroidal magnetic field [2]. In that case the analysis was made on the main components of ITER relevant samples introduced in the FTU vacuum vessel, flush with the first wall, by the Sample Introduction System. In this paper results of LIBS measurements, carried out from remote (~ 2.5 m) on the FTU Mo (TZM) toroidal limiter tiles, during machine maintenance, are reported. The aim of this work was the measurement of their surface elemental composition, and in particular of the retained deuterium (used as a proxy for tritium).

2. Experimental layout

The FTU tokamak [3] is provided with an inboard TZM (0.40-0.55 wt% of Ti, 0.06-0.12 wt% of Zn, 0.06 wt% max of others metals, Mo balance) toroidal limiter, consisting of twelve sectors, each one supporting 30 tiles (see Fig. 1). The LIBS experimental lay-out, located out of an equatorial port, is visible in Fig. 2. A Quantel laser "Twin BSL" (λ = 1064 nm) has been used to probe, through the 2 inches sapphire window of the port, different points of the limiter section accessible from outside (see Fig. 3). The laser is able to deliver a double pulse, but the limited available experimental time allowed only the single pulse technique to be used. Collinear transmission of the laser beam and detection of the visible lines emitted by the LIBS plasma was done by using a dielectric mirror. The light emitted by the plasma plume generated by the laser pulse was focused on a bundle of 12 optic fibres, with 100 μm core coupled with a Jobin Ivon "Triax 550" spectrometer (550 mm) with 2400 grooves/mm grating. The latter, allowing for about 10 nm to be explored, was used to get the best spectral resolution to distinguish the D_α line (λ = 656.11 nm) from the H_α line (λ = 656.29 nm). The spectroscopic LIBS signal was acquired with an Andor "Istar DH320T-18F-63" ICCD camera with

1024x512 sensor (26 um pixels). Laser and optics for laser transmission and visible lines detection were mounted on a plate movable along three axes and able to be pivoted, in order to sample as large as possible surface of the limiter through the narrow and long port. The laser spot size, as approximately inferred from the remore visualization of the crater dimensions, was about 5 mm^2; it corresponds to a power density of 0.4 GW /cm^2. This power density value, although not very large, still satisfies the requirement of stoichiometric ablation for all possible surface components, including molybdenum: for the latter the minimum laser intensity I_{min} needed to produce vaporization is just about 0.4 GW /cm^2, calculated according to Moenke-Blakenburg formula [4]:

$$I_{min} = \rho L_v k^{1/2}/\Delta t^{1/2} \text{ (W/cm}^2) \qquad (1)$$

where ρ is the material density, L_v the vaporization heat, k the thermal diffusivity and Δt is the laser pulse duration. Timing and duration of the LIBS measurements, carried out when FTU was not operating, had to comply with the schedule of machine shutdowns. Measurements under vacuum (including after a boronization) and with nitrogen and argon atmosphere have been done.

3. Results and Discussion

3.1 Under vacuum measurements

As shown in Fig. 4, measurements carried out under vacuum resulted in a good resolution of D_α (656.1 nm) and H_α (656.28 nm) lines. In the same spectra five lines of Ti I have been identified: Titanium is present in TZM with a very low percentage (about 0.5 wt%) so that the appearance of these lines could be the result of some segregation of this element to the very surface of the tile. On the other hand, as will be discussed later in the paper, attempts to quantify the surface elemental concentration by LIBS were prevented by the narrow detected wavelength range and the not homogeneous surface composition of the tiles. Deuterium and hydrogen were also detected and well resolved in shadowed zones in between the tiles (see red arrows in Fig. 3), about 1 cm under the plasma facing surfaces (Fig 5).
Depending on when measurements was done, also spectral lines of other elements (mainly Li, B , Mo) have been identified, by changing the explored wavelength range. In Fig.6, showing a spectrum obtained close to the period of liquid lithium limiter experiments [5], inserted in the FTU vacuum vessel through a vertical port distant about 1 meter, strong Li I line is visible. Molybdenum lines have been detected usually after some laser shots on the same point, Mo being the main constituent of the TZM tiles.
Right after a boronization of the vacuum vessel [6] (routinely performed in FTU), carried out by using a dc-glow discharge in a throughflow of 90% of helium and 10% of deuterate diborane (B_2D_6) and depositing a boron layer about 100 nm thick, LIBS measurements with reduced laser energy were carried out in the very surface layer of a tile to detect Boron and Deuterium lines. The first three spectra in Fig. 7, corresponding to subsequent laser shots on the same point, show the B I line, then disappearing in the next shots. In a laser spot close to the one showing B I lines, Deuterium was also detected together with Hydrogen (Fig. 8). From the Voigt fit of Dα / Hα the Gaussian and Loretzian contributions to the line profile have been retrieved, so that the electron temperature and density of the LIBS plasma were estimated to be $T_e = 1.1$ eV, $n_e = 7.2\cdot10^{15}$ cm^{-3}, respectively. According to the McWhirter criterion [7] the n_e value is close to the lower limit of electron density necessary for the existence of Local Thermodynamic Equilibrium (LTE). Besides this, the narrow wavelength window of the spectrometer, optimised for high resolution power, required several laser shots to detect the spectral lines of all surface components; by taking into account that the tile surface composition is not so homogenous as usually it is for laboratory samples and that it was needed to avoid the drilling of too many craters on the tiles, Calibration Free (CF) method [8] was not applied to these, and to all other spectral data, for quantifying the elemental concentrations. An alternative approach, although affected by not negligible errors in these a little marginal LTE conditions, is to obtain the concentration ratio of each of the detected elements with respect to one of them having significant emission lines in the wavelength regions under study. This element is then considered as an internal standard. By applying this method with D as internal standard the relative concentration of H and B respect to D were obtained as: $[D]/[H] \approx 2.7$, $[D]/[B] \approx 3.1$ the latter resembling stoichiometric composition of deuterated diborane. As other contaminants were not present in the thin explored layer, the closure relation ([D] + [H] + [B] = 1) could be applied and the absolute atomic concentrations of the three elements could be obtained as: [D] ≈ 59%, [H] ≈ 21.9%, [B] ≈ 19.1%. On the other hand the CF capability to provide quantitative results was well assessed in many laboratory experiments [9], so it is to be expected that more suitable experiment parameters and the simultaneous use of a broadband spectrometer (detecting many emission lines) will enable CF method to be use to quantify elemental concentrations in the foreseen LIBS measurements with a robotic arm on FTU First Wall.

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3.2 Measurements in Nitrogen and Argon atmosphere

Measurements with Nitrogen atmosphere (10^5 Pa) led to negligible hydrogen and deuterium signals. A possible explanation could be the not suitable setting of detection parameters as the result of limited available time. Besides this a role could be played by the partial formation of ND compounds [10], of which the emission lines in the LIBS plasma plume, at 335.7 and 336.4 nm, were not detectable in our experiments given the cut of wavelengths below 400 nm, caused by the dielectric mirror. The signals coming from D and H not chemically bound and/or from the brake-up of compounds could be also below the detection limit. The unsuccessful measurements with N_2 atmosphere, to be confirmed, indicate that it might be a limitation with nitrogen to be used in ITER for LIBS studies. Argon measurements (5×10^4 Pa) resulted in deuterium and hydrogen lines well visible (Fig. 9) although with worse resolution with respect to vacuum measurements, as it was to be expected because of the larger Stark broadening of emitted lines.

4. Conclusions

The capability of detecting from remote, with good resolution, the deuterium retained in the tiles of the FTU toroidal limiter has been demonstrated under vacuum conditions. Deuterium codeposited with boron as a result of wall conditioning by boronization using deuterated diborane, has been detected. This supports the planned strategy in the frame of WPMST2-20 project (LIBS measurements with a robotic arm, jointly with CU, FZJ, IPPLM, LU, UT, VTT associations), with the use of deuterated boron film in the FTU tokamak to simulate the situation foreseen in ITER, where codeposition with beryllium will be the main cause of tritium retention in the machine.

Libs measurement with nitrogen and argon atmosphere resulted in negligible presence and in not well resolved Dα and Hα lines, respectively. To be noted that in this case, as the result of limited available time, the optimization of the experiment parameters was not as good as in vacuum conditions and that Double Pulse technique, a promising tool to be used in atmospheric conditions, was not used [9].

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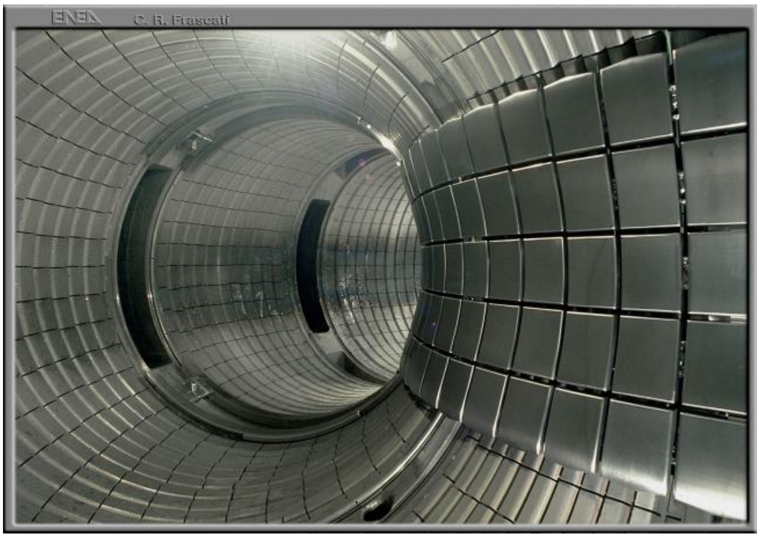

Fig. 1 – FTU vacuum vessel with inboard toroidal limiter

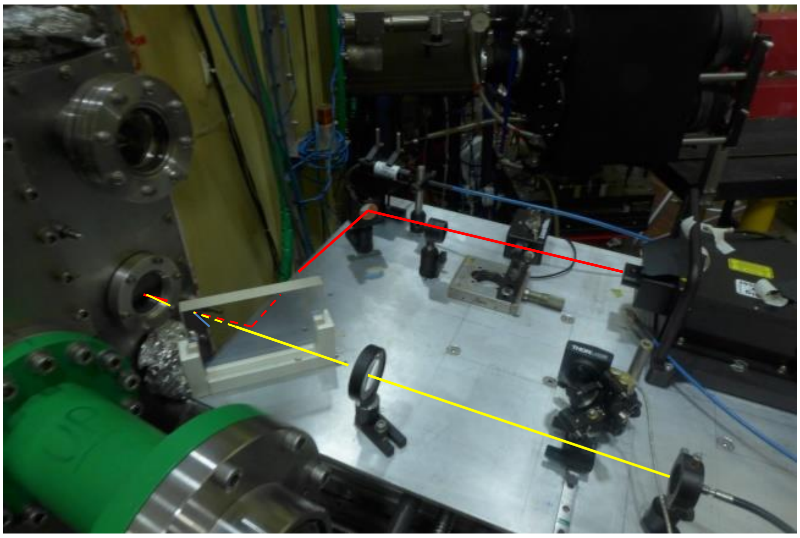

Fig.2 – Photo of the LIBS experimental lay-out (red line: laser path, yellow line: visible emitted light)

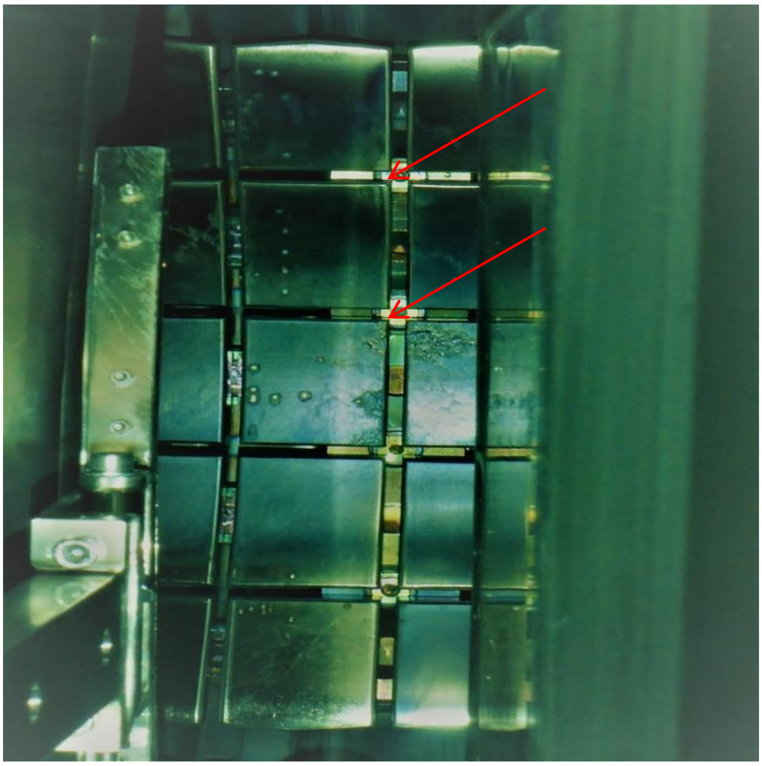

Fig. 3- View of the toroidal limiter sector as seen from the equatorial port. Craters by laser shots are visible on the tiles. The red arrows indicate the shadowed part analysed

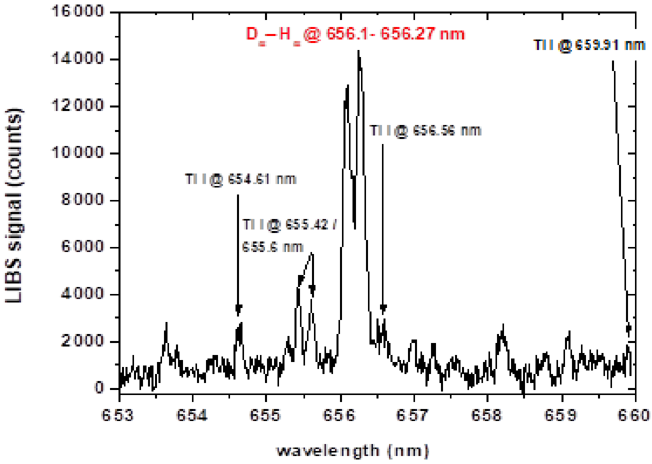

Fig. 4 – Under vacuum LIBS spectrum showing good resolution of D_α and H_α lines

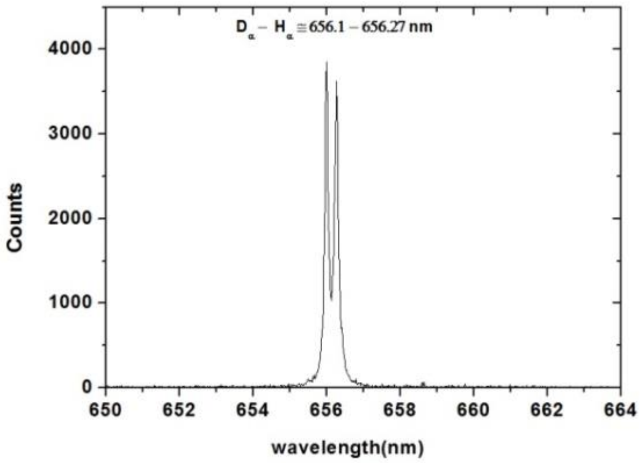

Fig. 5 – Under vacuum LIBS spectrum detected on a point shadowed by adiacent tiles

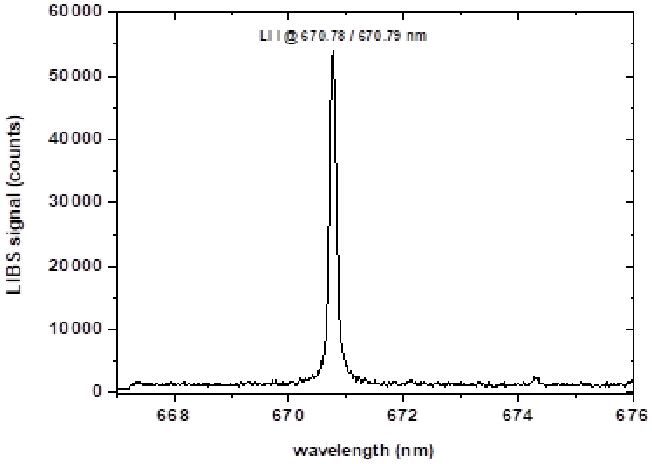

Fig. 6 – Under vacuum LIBS spectrum, detected after a period of Liquid Lithium Limiter experiments, showing an intense Li I line

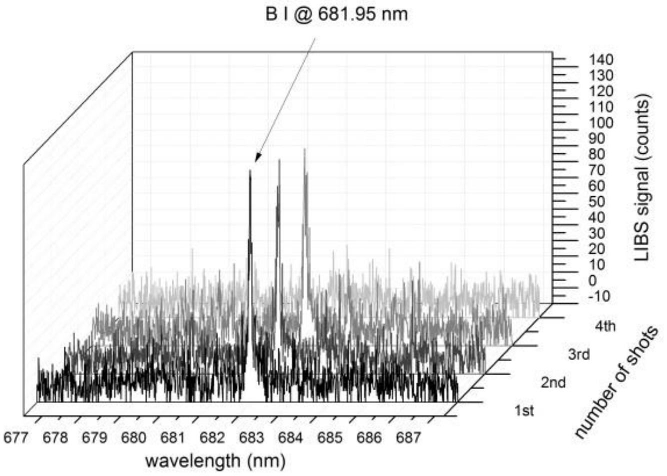

Fig. 7 – Series of three spectra detected just after the vacuum vessel boronization, showing the presence of B I line.

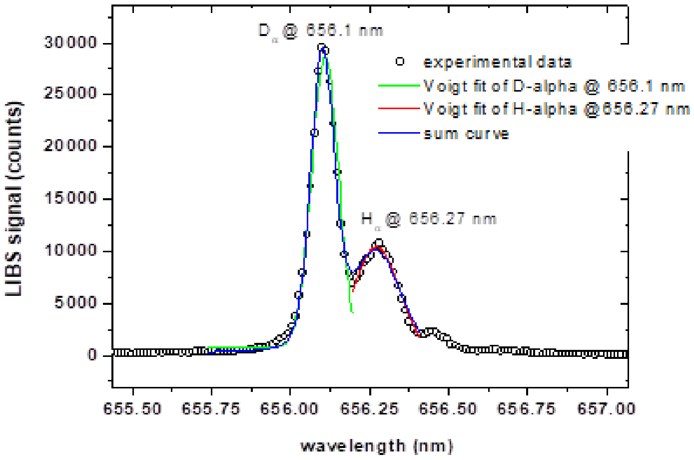

Fig. 8 – Under vacuum spectrum of D_α and H_α lines just after vacuum vessel boronization

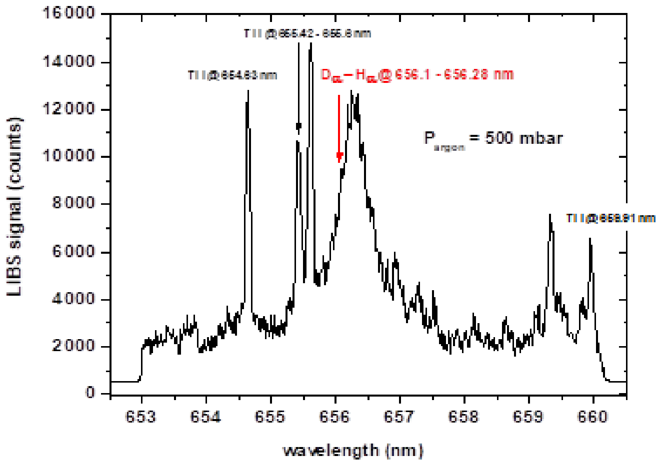

Fig. 9 – LIBS spectrum, detected in Ar atmoshere (500 mbar) showing D_α and H_α lines not well resolved. Titanium lines are also visible.