Pulsed laser removal of tungsten nanoparticles: surface analysis and visualization of particle ejection dynamics

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Abstract

In this study, we report on some of the aspects that describe the interaction of nanosecond UV laser pulses with solid surfaces covered by layers of tungsten nanoparticles aggregates, with an interest in applications for next-generation fusion reactors. The safe and efficient operation of such reactors is challenged by their unavoidable contamination with dust by-products due to interactions between plasma and the surface of plasma-facing components. We investigate the cleaning possibility of tungsten nanoparticle layers from solid surfaces using a KrF excimer laser source with tailored spot shape. The samples that emulate dust contaminated surfaces are synthesized by magnetron sputtering combined with gas aggregation technique. In order to shed light into the particle removal efficiency, the tungsten samples are investigated by scanning electron microscopy, Raman spectroscopy, X-ray diffraction, and profilometry. Furthermore, in order to gain insight into the W nanoparticles and fragments mobilization dynamics, a plasma imaging setup is used to visualize the speed and directionality of the ejected material. We find that a very good surface cleaning can be achieved, even for the lowest considered laser fluence (0.5 J/cm²) following 8 pulses. Material ejection from the laser irradiated surfaces occurs in two regimes, over very diverse time scales: i) first, a fast plasma plume travelling at 40,000 km/h reaches the substrate in a few microseconds, ii) followed by the ejection of large particle conglomerates at speeds of the order of few hundred km/h. The particle ejection dynamics is shown to differ depending on the type of atmosphere (ambient pressure and vacuum, respectively), whereas the plasma plume evolution could only be evidenced under vacuum conditions.

Keywords: tungsten nanoparticles, laser cleaning, plasma dynamics
1. Introduction

The sheer complexity of next-generation fusion reactors, such as the International Thermonuclear Experimental Reactor (ITER) and ITER’s successor DEMO (DEMONstration Power Station), will raise numerous issues related to their safe and efficient operation. These large-scale tokamak-type devices are being designed based on the idea that a clean and cheap energy can indeed be harnessed through an intricately tailored and controlled fusion reaction [1]. It is beyond the scope of this paper to refer to all the issues that have so far hindered or delayed the successful operation of such fusion reactor concepts. Instead, our study relates to one particular problem that is expected to arise and to negatively affect the safety and efficiency of next-generation fusion reactors, i.e., their unavoidable contamination with dust by-products due to interactions between the plasma and the surface of plasma-facing components.

Exposure to plasmas has been shown to produce damage features particular to tungsten surfaces. This damage manifests itself in very different ways, which have been shown to strongly depend on the experimental conditions, quality of tungsten tiles, reactor geometry and many other factors. Still, most of these damage types are collectively referred to as: defect accumulation, erosion, fuzz, blisters/bubbles etc. [2, 3, 4, 5, 6] Plasma-wall interactions represent a problem that continues to hinder the realization of fusion reactors demonstrating energy generation, which results from the fact that the fusion process gives rise to high quantities of dust powder (micro and nanoparticles of tungsten, as well as carbon, beryllium, etc.) due to an erosion of the vacuum vessel, especially in the divertor region [7, 8]. These particles may incorporate important amounts of tritium, and are transported in remote areas and in gaps present in the in-vessel topology. Tritium accumulation in the dust deposits would also compromise the safe operation of fusion machines.

From an applicative point of view, a solution needs to be found to address the surface contamination of reactor components with tungsten particles. The possibility of their removal and/or mobilization from various types of surfaces using laser sources is something that has already been considered. [9-Error! Reference source not found.] However, in the absence of actual samples that have been subjected to plasma fluxes to be produced in next-generation fusion reactors, the research groups involved in the study of laser particle removal from contaminated surfaces have resorted to using various types of self-made samples that emulate such surfaces. For instance, Vatry et al. [9-Error! Reference source not found.] have used pulsed laser
deposition (PLD) to obtain surfaces covered with highly adhesive tungsten nanoparticles, but the actual density of these particles on the considered surfaces was extremely low, with no interaction between them. This is very different from the case of a real fusion reactor, such as ITER, where large quantities of dust by-products are expected to be produced through plasma-wall interactions. In another study, Stokker-Cheregi et al. \cite{11} have obtained surfaces covered by tungsten nanoparticles by the means of pulsed laser ablation of bulk tungsten targets immersed in different liquids. Thin films of tungsten nanoparticles were obtained on a substrate following the evaporation of the liquid, but this approach also proved to have some drawbacks, such as the fact that it is difficult to control the thickness of the nanoparticles thin layer, or their distribution on the covered surface.

In terms of dust removal techniques, it should also be mentioned that different methods have been proposed. For example, A. Hakola \cite{12} proves that arc-discharge and plasma-sputtering methods are efficient at cleaning the dust, but the feasibility of applying it to the ITER vacuum vessel is debatable due to fusion reaction requirements and vacuum vessel geometry. The pulsed laser ablation/cleaning technique is conceptually very simple, especially since it has already proven its efficiency in artwork restoration projects, where high sensibility and selectivity are needed \cite{13,14}. This technique has many advantages such as: versatility (many materials can be ablated), fast cleaning of contaminated surfaces, suitability for vacuum conditions, cost-effectiveness, eco-friendliness, and ease of automation.

Therefore, in this work we have addressed some of the issues mentioned previously by studying the laser removal of tungsten nanoparticles in the form of dense layers, which have been obtained by a technique that combines magnetron sputtering with gas aggregation (MS-GAS) \cite{15}. The MS-GAS technique has proved to be a fast and inexpensive technique to obtain relatively large-sized films, having diameters of several mm, of dense tungsten nanoparticles aggregates. This achievement has given us the opportunity to carry out particles removal experiments that are relevant in terms of practical applications in which the size of the contaminated area and density of contaminants are the most important issue to tackle. The novelty of our approach stems from the fact that this is the first time that the laser removal of such dense tungsten nanoparticles layers was investigated in terms of efficiency. To this end, the particles removal experiments were carried out using a KrF excimer laser operating at 248 nm wavelength and 1 Hz repetition rate. The advantage of such a laser system is the fact that it
exhibits a flat beam profile, rather than a Gaussian one that is typical to solid state high-powered pulsed lasers, which allows for its shaping at the focusing position over areas up to $1 \times 1$ mm in size, while at the same time preserving the homogeneous intensity distribution of the beam profile. The sample surfaces were analyzed using scanning electron microscopy (SEM), Raman spectroscopy, X-ray diffraction and profilometry. Moreover, we used a plasma-imaging setup to visualize the speed and directionality of the luminescent large particles ejected following laser interaction, both in ambient atmosphere and in vacuum. By employing this approach, we are able to gain deeper insight into processes that are relevant for future decontamination applications.

2. Experimental

2.1. Fabrication of tungsten nanoparticle layers

The tungsten nanoparticles (W NPs) films are obtained by magnetron sputtering combined with gas aggregation from a bulk tungsten target. The experimental setup has two major components: a deposition chamber and a MS-GAS cluster source (aggregation chamber), linked through an aperture with a diameter of 2 mm. The tungsten target is placed directly in front of the magnetron sputtering device at a distance of 90 mm inside the cluster source. The process gas is fed into the system through the aggregation chamber. Pure Ar (99%) is used as process gas, with 5 sccm flow rate that corresponds to a pressure of 80 Pa in the aggregation chamber and a pressure of 0.5 Pa in the deposition chamber. This difference in pressure is due to the fact that the system is pumped down with a turbomolecular pump attached to the deposition chamber, whereas the two chambers are connected through a very small aperture. The tungsten vapors are supplied to the clustering process via a sputtering process. The sputtering process is run by a radio frequency power supply (RF, 13.56 MHz) operating at 60 W. The resulting tungsten nanoparticles are collected on Si (100) mounted inside the deposition chamber at a distance of 30 mm from the aperture. The collection time is 30 minutes, and the final thickness of the W NP layers is of the order of few microns. Details on the tungsten nanoparticle experimental growth process can be found in our previous work \[15\].

2.2. Laser based system for tungsten nanoparticle removal
Previous studies have shown that lasers working in the UV range are much more efficient at the removal of tungsten nanoparticles from flat surfaces than lasers operating in the IR range \(^{[16]}\). Therefore, in this work, all tungsten nanoparticle removal experiments are carried out with a KrF excimer laser (Lambda Physik LPX 300, 20 ns pulses) operating at 248 nm wavelength and 1 Hz repetition rate.

A scheme of the laser removal experimental setup is shown in Fig. 1. In order to carry out the laser removal experiments, the substrates with W NP layers are placed on a computer-controlled translation stage, which allowed their continuous displacement, and irradiated with the 248 nm laser beam (from the front side). The angle between the laser beam and the thin film surface is kept constant at 90°. A mask is used to image a rectangular laser spot (1 × 1 mm) with a top-hat beam profile on the surface of the W NP layers. The experiments are carried out at different laser fluences: 0.5, 1, 1.5, and 2 J/cm\(^2\), respectively. The material ejected from the W nanoparticle layers following their laser irradiation was collected on a silicon substrate placed opposite to the thin film surface at a distance of 4 cm. Experiments involving both laser removal and collection of ejected material have been carried out at ambient temperature, whereas plasma imaging analyses were also carried out under vacuum (1×10\(^{-4}\) Pa pressure).

2.3. Morphological, structural, and compositional analysis of the W NP layers

X-ray measurements of the as-deposited W NP layers are carried out in a Bragg-Brentano geometry (θ-2θ, between 20° and 90°) using a Siemens D500 powder diffractometer equipped with a Cu-source with 0.1 mm wide slits.

The Raman spectra of the as-deposited W NP layers are recorded on a Labram confocal Raman microscopy system from Jobin Yvon. The excitation source is the 633 nm line from a HeNe laser. For each measurement, the exposure time and the number of accumulations were 10 seconds and 5 times, respectively. Raman spectra are collected over the range of 200-1000 cm\(^{-1}\), at room temperature. Both X-ray diffraction and Raman measurements were carried out on the as-deposited W layers, prior to their laser irradiation experiments.

The distribution of the W NPs on the substrates after their deposition and following laser removal experiments is investigated by scanning electron microscopy. The images are obtained from top view.
analysis and are acquired with a Zeiss Supra VP55 FE-SEM apparatus operating at a voltage of 5kV and using an in-lens detector.

Profilometry is used to measure the depth of the laser induced craters on the surface of the W NP layers. The depth profiling measurements are carried out with a Dektak 8 profilometer from Veeco. The ablation depth is measured by averaging the profile of the crater.

2.4. Plasma imaging

The evolution of the ejected material is investigated by plasma imaging to gain information on particle trajectories and velocity. An intensified CCD camera (Andor i-Star) is used to record the time evolution of the ejected material. The camera is triggered externally by the laser pulse using a photo-diode. The on-board digital delay generator is used for gating the image intensifier (typically with a gate of 100 ns), and increasing time-delays are used to capture the time evolution of the ejected material. A more detailed description of our plasma imaging technique and setup can be found in [17].

3. Results and discussion

Our present study capitalizes on the previous success we had in obtaining tungsten layers of aggregated nanoparticles [15], which opened the possibility of carrying out dust removal experiments under experimental conditions relevant to those to be encountered in next-generation fusion reactors. Our results are presented and discussed in view of achieving a comprehensive picture of both: i) surface properties from a structural and morphological point of view, which present a broader fundamental appeal that can extend beyond the immediate applicative scope of this research; and ii) material ejection kinetics due to laser-surface interaction.

SEM investigations carried out prior to laser irradiation have revealed that the surface of our samples is covered with aggregated nanoparticles formations, as shown in Fig. 2a and 2b. The W NPs have a flower-like aspect and are evenly distributed on the surface of the substrate. The surface of samples has no major defects, such as cracks and crannies. The diameter of individual W NPs is between 70 and 200 nm, with an average size of ~100 nm. The cross-section image of a W NPs layer is shown in Figures
2c and 2d. The W NPs form a micron-sized thick compact layer on top of the silicon substrate, with no cracks along its thickness.

The X-ray diffractogram shows the presence of a crystalline phase and of a disordered, almost amorphous phase. The α-W phase corresponding to the (110), (222), (420), (421) reflections and the β-W phases centered around (210), (210) peaks, alongside with the peaks coming from the silicon substrate (marked with "*") are identified in Fig. 3a. Tungsten is known to exists predominantly in two crystalline forms: α and β. In our X-ray diffractogram, the dominant crystalline phase is α-W. No information regarding the presence of crystalline tungsten oxides could be extracted from the X-ray diffractogram. However, this does not exclude the presence of tungsten oxides in our samples. It is possible that tungsten oxides are present in the amorphous phases or the thickness of the oxide layer is not sufficient to give an X-ray reflection signal.

The XRD pattern of the as deposited W nanoparticles (W-NP) layer is shown in Fig. 3a. The peaks match the β-W (A15 structure) standard, card no. 47-1319, from the International Center of Diffraction Data (ICCD) database. The standard pattern is depicted at the bottom of Fig. 3a, and the pattern is Miller indexed accordingly. From the two allotropic forms of W, the β-W phase, having the topologically close-packed A15 structure (space group Pm-3n) is the meta-stable one, while the α-W, which is body-centered cubic (A2, space group Im-3m), is the thermodynamic equilibrium stable phase [18,19]. The XRD pattern of our as deposited W NP layers reveals a pure β-W phase, with no α-W reflections, indicative of a stabilization of the β-W phase. The growth and the stabilization of the β-W film has been previously explained essentially by the oxygen impurities incorporated during the film growth [20,21]. Although in our case no information regarding the existence of crystalline tungsten oxides could be extracted from the X-ray diffractograms, likely due to the lack of long-range order, Raman analysis of the as-deposited W layer presented in Fig. 3b (red line) indicated their presence. The Raman spectrum of the W NPs thin film, acquired in the 200-1000 cm⁻¹ range, shows four vibrations bands centered at 271, 326, 714 and 806 cm⁻¹. In previous studies, the peaks centered around the 806 and 714 cm⁻¹ wavenumbers have been assigned to crystalline WO₃ (m-phase), and correspond to the stretching vibrations of the bridging W–O–W [22-25], more specifically attributed by literature to W-O stretching (ν), W-O bending (δ) and O-W-O deformation (γ) modes [22-25]. The 271 and 326 cm⁻¹ peaks
correspond to the bending vibration $\delta$ (O-W-O) [22-25]. The high intensity of the peak at 271 cm$^{-1}$ indicates that a great fraction of crystalline phase is present in our samples, this peak being also a label for the presence of crystalline WO$_3$ [22-25].

Furthermore, Raman measurements have been carried out on our samples after laser irradiation of the W layers at a laser fluence of 0.5 J/cm$^2$ and different numbers of pulses, as displayed in Fig. 3b. The position of the silicon peak at 523 cm$^{-1}$, used for spectrum calibration, is in good agreement with the literature [26]. These measurements reveal the progressive removal of the tungsten layers with increasing the number of laser pulses from 7 to 10, in agreement with the profilometry analysis discussed in the following.

The line profile, from profilometry analysis, of a W NP layer after laser irradiation with different numbers of pulses is presented in Fig. 4. The removal process appears as inefficient after applying between 1 and 4 laser pulses. The only obvious effect after 1-2 laser pulses is a surface melting followed by re-solidification of the top-most layers, whereas after 3-4 laser pulses the film also begins to crack (see SEM images from Fig. 5). Essentially, only a change in surface morphology is observed by applying between 1 and 4 laser pulses, but no removal of material. The first signs of material removal from the film are seen after 5 laser pulses, whereas after 8 laser pulses and by applying a laser fluence of 0.5 J/cm$^2$ the W NPs are almost completely removed, without any damage to the silicon substrate, as can be seen from Fig. 5e and f. This indicates that the mechanical strength of the bond between the laser-irradiated material and non-irradiated areas needs to decrease before large fragments of NP agglomerates can be displaced by subsequent laser pulses.

The SEM image of the ejected material collected on parallel Si supports is presented in the inset in Fig. 5f. After laser irradiation (at a fluence of 1 J/cm$^2$), it can be seen that some of the collected W NPs are melted and fused around the remaining unmelted W NPs. The size of the collected W NPs agglomerations ranges from 20 to 50 µm.

The subsequent plasma imaging experiments were envisioned in order to check this apparent result that the material from the initial thin films is extracted in the form of sizeable clusters, rather than as individual particles. By carrying out plasma imaging measurements (in air and in vacuum, respectively),
we evaluated the particle dynamics of the W NPs thin film fragments following laser irradiation. It should be noted that, in the Fig. 6 to 8, the images of particle dynamics acquired at different delays correspond to different laser pulses applied to different positions on the surface of the samples, and therefore they do not represent a continuous motion capture following a single laser pulse at different time delays. This is also the reason why some discrepancies in terms of particles distribution may appear at different time delays in the same set of images.

As illustrated in the top image of Fig. 7 (i.e. 'Sample position'), the target samples are placed on a support on the left-hand side of the image (with the surface perpendicular to the plane of observation), whereas the ablated particles reach the reference surface to the right-hand side of the image, where a typical substrate mounting support can be seen. The distance between the target sample surface on the left side and the reference surface on the right side is 4 cm. Both of the target sample and reference surface are also visible in Fig. 6 at short delay (1 µs), as the intense laser pulse undergoes strong scattering. The particle velocities are estimated from the distance travelled by the particle front relative to the time (i.e. image acquisition delay) taken to traverse that distance.

The time evolution of tungsten NP layer fragments irradiated in ambient atmosphere, with the irradiated surface of samples on the left side of the images and the mobilized W NPs layer fragments travelling to the right, is shown in Fig. 6. During the ablation experiments, highly luminescent particles were visible by naked eye, as a result of their thermal continuum emission. Therefore, the intensity of light was considerable, and enabled an optimum imaging with gate times of the order of ~100 ns, a time window two orders of magnitude smaller than the actual delay between the captured frames, which was of 80 µs. Fig. 6a reveals the trajectory of the particles ablated at a laser fluence of 0.5 J/cm², whereas in Fig. 6b the laser fluence was set at 1 J/cm². Figure 6b reveals a marked fragmentation of the ablated material, and consequently a broader distribution of particles velocities, as compared to Fig. 6a. The particle front in Fig. 6b also has a much more elongated shape, although the foremost particles travel about as fast as those in Fig. 6a. Although there is a factor of two between the two considered fluences, the velocities of the ablated fragments in air have quite similar values, i.e. ~110 km/h.
The time resolved images of the W NPs samples irradiated in vacuum are displayed in Fig. 7. The experimental setup and process parameters are the same as before, but the tungsten film is now in vacuum \(1 \times 10^{-4}\) Pa. The ejected W NPs thin films fragments show an increased angular dispersion with respect to ambient atmosphere conditions, which was to be expected when conducting the same experiments under vacuum conditions. It should also be noticed that in this case the exposure time was increased to 1 µs. The general finding is that with increasing fluence, the velocity of the nanoparticles also increases, as well as their amount. At a fluence of 2 J/cm\(^2\), the particle front reaches the surface to the right in less than 400 µs. A remarkable number of nanoparticles are reaching the right-side surface, and a lot of them can be seen bouncing back from it. Concerning velocities, the first 80 µs show a very fast expansion, after which a steady velocity is achieved until the nanoparticles reach the reference surface. The values of these velocities are displayed in the last column of Fig. 7. Once again, it is important to mention that each image corresponds to one independent laser shot and we noticed some variability of ejected species between each shot, which means that there may be some uncertainty in the calculation of the respective velocities.

Another aspect that is noticeable at short delays in Fig. 7 is a plasma plume. In order to better resolve this feature, we also captured the expansion of the plasma plume for the fluence of 1 J/cm\(^2\) using reduced exposure times (exposure of 50 ns) and shorter time steps (delay of 500 ns). The results are displayed in Fig. 8, and they show that after the initial 0.5 µs expansion, the plasma expands at an average velocity of 40,000 km/h, reaching the reference surface in ~2.7 µs, which is two orders of magnitude faster than the W NPs thin film fragments.

We point out the fact that the images presented in Fig. 6 to 8 are not spectrally resolved, but they are actually a focal-plane projection of the experimental setup onto the sensor of the ICCD camera. The detected light emitted by the visualized particles comes as a result of: i) thermal continuum emission following excitation by the intense laser pulses, in the case of ejected tungsten particles/fragments in Fig. 6 and 7, or ii) as radiative recombinations in the case of the fast plasma plume shown in Fig. 8.

It has been suggested that the removal mechanism of tungsten nanoparticles is linked to photoelectrons and to thermal degradation of the particle aggregates [9, 10]. As such, thermal induced stress is a likely
factor which would cause a mechanical fracture in the tungsten nanoparticles layers, as suggested by the cracks appearing in Fig. 5. The photoelectrons extracted from the tungsten nanoparticles could generate negative oxygen ions from surface WO$_3$ compounds evidenced in Raman spectroscopy measurements, which in turn give rise to an electrostatic force greater than the adhesion forces between the positively charged nanoparticles and the negative species contained in the volume above it [10]. In our experiments, the removal process is assigned to a complex mechanism involving the ejection of fast ions/neutral atoms in a plasma phase, followed by the ejection of large "hot" fragments of micron-size agglomerated NPs from the thin film layer, as supported by the SEM investigation performed on the collected fragments (see inset in Fig. 5f).

4. Conclusions

We have successfully demonstrated the removal of tungsten nanoparticles from compact layers with a KrF excimer laser operating at 248 nm. Our approach allowed us to make significant steps towards resolving a realistic situation in ITER-like fusion reactors. Due to the unavailability of actual reactor components that had been subjected to plasma interactions, our proposed approach relied on obtaining thin films of dense tungsten nanoparticles aggregates using the MS-GAS technique. One of the most important achievements of our approach was the removal of W NPs conglomerates without damaging the underlying substrate surface. This result could be of practical importance not only for dust removal applications in next-generation fusion reactors, but also for applications in microelectronics engineering. Given the tungsten tile-based design of the divertor region of current ITER-like fusion reactors, a laser treatment of such surfaces can be implemented as an 'ex-situ' solution for damaged/contaminated tiles that have been disassembled and removed from the reactor, and need to undergo reconditioning prior to subsequent reassembly.

From a fundamental point of view, we have evidenced for the first time the dynamics of the mobilized W NPs conglomerates under ambient atmosphere and vacuum conditions, respectively. The ejection of material from the laser-irradiated surfaces was found to occur in two regimes, over very diverse time scales: i) a fast regime, of few $\mu$s, attributed to plasma plume expansion; and ii) a slow regime, occurring over several hundreds of $\mu$s, in which the ejection of tungsten particles/fragments takes place. We have
computed the velocities of the ejected particle conglomerates to be of the order of few hundreds of km/h, and we also evidenced important differences in the divergence and spread of the particles front, depending on the laser fluence, a result which will be of interest when considering the development of future in-situ dust collection solutions.

Acknowledgements

Financial support from i) NILPRP, through the LAPLAS VI NUCLEU program no. 16N/08.02.2019 ii) the EUROfusion Consortium under work-package WPENR from the European Union’s Horizon 2020 research innovation program iii) the Romanian Research Ministry under contracts 1EU-1/2 / 2016, IFA-CEA No. C5-07 / 2016 iv) and (1) the TRANSAT project funded by the Euratom Research and Training Programme 2014–2018 under grant agreement No 754586 are gratefully acknowledged.

We are thankful for the contribution of Dr. Ruxandra Birjega in helping us interpret crystallographic data.
**Figure caption**

**Figure 1.** Scheme of the experimental setup for tungsten nanoparticle removal.

**Figure 2.** (a) and (b) Top view SEM images, and (c), (d) cross-section SEM images taken at different magnifications of an as-deposited W NPs layer.

**Figure 3.** (a) X-ray diffraction pattern of the as-deposited W NP layer; (b) Raman spectra of the as-deposited W NP layer, Si substrate (* indicates peaks assigned to Si), and W NP samples after laser irradiation at 0.5 J/cm² laser fluence and different number of laser pulses.

**Figure 4.** The line profile obtained from profilometry measurements of our samples after laser irradiation with different number of pulses (between 1 and 10). The laser spot size is 1 mm × 1 mm and the laser fluence is 0.5 J/cm².

**Figure 5.** Top view SEM images of a W NP layer irradiated with a) 2 laser pulses, c) 4 laser pulses, and e) 8 laser pulses. b) Top-view SEM image of a W NP layer irradiated with 2 laser pulses, showing that W NP nanoparticles on top of the irradiated surface have melted. d) Top-view SEM image of a W NP layer irradiated with 4 laser pulses, in which cracks may be noticed in the irradiated W NP layer, f) Top-view SEM image of the edge of a W NP layer irradiated with 8 laser pulses and (inset): SEM images of tungsten film fragments collected following their ablation from the target. The laser spot size is 1 mm × 1 mm and the laser fluence is 0.5 J/cm².

**Figure 6.** Time resolved images of the ablated W NPs thin films fragments ablated at ambient pressure, at fluences of (a) 0.5 J/cm² (B/W scale) and (b) 1 J/cm² (green color gradient), respectively.

**Figure 7.** Time resolved images of the W plasma plume expansion in vacuum at long delays and 1 µs exposure time, for laser fluences between 0.5 and 2 J/cm².

**Figure 8.** Time resolved images of the W plasma plume expansion in vacuum at short delays and 50 ns exposure time, for a laser fluence of 1 J/cm².
Figure 2
Figure 3
Figure 4
Figure 5
Figure 6
### Figure 7

#### Vacuum [exposure of 1µs]

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Figure 8

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**Vacuum [exposure of 50ns]**

4 cm
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