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Executive Summary of the Thesis

Production of Novel Nanostructured Targets for High-Intensity Laser-Plasma Interaction Experiments

Laurea Magistrale in Nuclear Engineering - Ingegneria Nucleare

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1. Introduction

Laser-plasma interaction is a field of study that has raised great interest in the scientific community. When a high-intensity laser interacts with a target material, this is rapidly ionized, generating a plasma. The continuous development of laser technology has allowed the exploration of various interaction regimes and phenomena that are relevant for fundamental research and possible applications. Laser beams delivering intensities $\geq 10^{15}\ W/cm^2$ in ns pulses are exploited in Inertial Confinement Fusion (ICF), one of the two approaches toward nuclear fusion energy production. At ultra-high intensities (i.e., $> 10^{18}\ W/cm^2$), particle acceleration can be achieved. In particular, TW laser-driven ion sources may provide a more compact and cost-effective alternative to conventional accelerators in various fields, ranging from materials science to medicine. To guarantee the feasibility of these appealing applications of laser-plasma systems, precise and reliable control of the interaction, together with an increase in the process efficiency, is of fundamental importance. To achieve these goals, along with advances in laser technology, proper target design and engineering is necessary. Indeed, the target material's properties influence plasma characteristics and strongly impact the laser energy absorption efficiency. Therefore, a lot of effort has been devoted to the development of advanced targets. The thesis work is devoted to the production of nanostructured targets with controlled and tunable properties for laser-driven ion sources and ICF. This is achieved through an interdisciplinary and synergistic approach, exploiting advanced deposition techniques, typical of material science.

2. State of the art on targets

2.1. Single Layer and Double Layer Targets for laser-driven ion sources

One of the major challenges for the development of laser-driven ion sources is to achieve energies suitable for the envisioned applications, exploiting currently available laser technology while tailoring target properties. The most reliable and understood acceleration mechanism is *Target Normal Sheath Acceleration* (TNSA) [1]. In its simpler configuration, a thin Single Layer target (SL) target, usually consisting in a free-standing solid layer, either polymeric or metallic, is irradiated with a ultra-high-intensity laser, generating a plasma. Plasma electrons interacting with the laser pulse are accelerated through the target and escape at the rear surface. There, a strong (few $MV/\mu m$) sheath field is generated due to charge unbalance. This is able to accelerate ions, mostly protons, up to a maximum energy of few tens of MeV. Protons are present at the rear surface of the target as impurities. It has been demonstrated that the energy of the accelerated ions increases with decreasing thickness of the SL target. Additionally, TNSA is dominant for target thicknesses ranging between hundreds of nm up to tens of μm, while for thinner films ($10 - 100\ nm$) other mechanisms such as Light Sail Radiation Pressure Acceleration might be more relevant, which may lead to higher achievable energies. Among the many proposed, a possible approach to increase the energy of the accelerated ions is to exploit Carbon based Double Layer Targets (DLTs), developed at the Micro and Nanostructured Materials Laboratory (NanoLab) of Politecnico di Milano [2]. Carbon foams are porous materials produced via Pulsed Laser Deposition (PLD), characterized by non uniformities at the nm and μm scale. Their density can be tuned to be close to the critical density ($n_c = m_e\omega^2/4\pi e^2$, being ω the laser frequency, e the electron charge, m_e the electron mass) which is few mg/cm^3 for the typical wavelengths of the employed lasers ($0.8 - 1\ \mu m$). In the DLT concept, the foam is deposited on the irradiated side of the solid foil to enhance laser-target coupling, thanks to the volumetric energy absorption allowed by the porous nanostructure. Commercially available thin foils can be used as solid layer in DLTs, although they present some limitations. Indeed, they are available in a limited number of thicknesses with an uncertainty in the nominal value up to 30%. Therefore, a hole-filling procedure was developed at NanoLab to realize metallic free-standing films with controlled properties directly on perforated target holders typically exploited in experiments. It is based on the filling the holes of the perforated holder with sucrose/caramel. Subsequently, the film (e.g., Titanium) is deposited via Magnetron Sputtering (MS) directly on the holder. Then, sucrose/caramel is dissolved in water, obtaining free-standing films in correspondence of the perforations. However, this process is quite time-consuming, only small-area free-standing films could be obtained, and the minimum thickness achieved was 200 nm.

2.2. Ablation layer in direct-drive ICF targets

In the direct-drive approach to ICF, a layered capsule containing the Deuterium-Tritium fuel is irradiated by a set of laser beams that begins to ablate the external surface, called ablation layer. The resulting plasma tends to expand outwards. Consequently, a shock is lunched through the capsule, compressing it until high temperatures and densities are reached, enabling the fusion reaction to occur [3]. The ablation layer plays a crucial role. Indeed, the overall implosion efficiency is strongly affected by the laser energy absorption efficiency of the ablator and by the occurrence of both hydrodynamic and laser-plasma instabilities, which are detrimental for the process. Therefore, a lot of effort is devoted to the improvement of the ablation layer performances. In particular, it has been proven that mid-Z ablators can mitigate laser-plasma instabilities. Light elements in the form of porous materials, such as chemically produced plastic foams, can increase the laser-target coupling. Indeed, thanks to their internal structure, foams are able to increase the laser absorption efficiency, smooth out laser inhomogeneities, and enhance the ablation loading [4]. Carbon nanofoams deposited via PLD are appealing materials to be used as ablation layers as they may combine the positive effects deriving from both mid-Z materials and porous structure. Nevertheless, while Carbon foams behavior has been widely investigated in the field of laser-driven ion acceleration, their behavior in ICF-relevant conditions is still an unexplored topic.

2.3. Objectives of the thesis work

The objective of the thesis work was to exploit the versatility of MS and PLD techniques to produce nanostrucutured targets with controlled and tunable properties according to envisioned application and laser parameters.

Specifically, concerning laser-driven ion sources, the aim was to develop a reliable *fishing* procedure to realize free-standing metallic films over a wide range of thicknesses. The fishing procedure can be summarized in four steps: I) realization of the sacrificial layer on the substrate, II) deposition of the films, III) film lift-off from the substrate exploiting a proper solvent and IV) subsequent fishing of the film with the perforated holder. These films may be used both in a SL or in a DLT concept. In this second case, the mutual compatibility between film and Carbon foam must be assessed.

In the frame of ICF, the goal of the thesis was to assess the feasibility of realizing Carbon nanofoam targets with suitable properties for laser-plasma interaction in ICF-relevant conditions.

3. Production and Characterization of nanostructured materials for targets

Since the realization of targets having controlled and tunable properties is of fundamental importance, the produced materials were characterized in terms of morphology and stresses, to evaluate how different deposition regimes and/or conditions affect their characteristics. Cu and Al are common target materials in laser-driven ion acceleration experiments. They were deposited via magnetron sputtering, either in Direct Current (DCMS) and High Power Impulse (HiPIMS) regimes. Carbon foams were deposited via fs-PLD for the realization of DLTs. ns-PLD was employed to realize C films and foams for the realization of targets for experiments in the field of ICF. The morphological characterization was performed by Scanning Electron Microscopy (SEM). Lastly, the surface curvature method allowed the evaluation of the average stress in Cu and Al films.

3.1. Cu and Al thin films

Cu and Al films were deposited via DCMS and HiPIMS fixing Argon (Ar) pressure to 0.5 Pa. The sputtering power was maintained fixed for all depositions at the following values: 547 W for Cu in DCMS, 295 W for Cu in HiPIMS, 506 W for Al in DCMS, 564 W for Al in HiPIMS. In the case of Al also biased (100 V) HiPIMS was exploited. The deposition time was tuned to obtain different thicknesses in the range 100 nm − 800 nm. Concerning Cu films, no substantial difference in the morphology of DCMS and HiPIMS deposited films could be observed. They showed a compact microstructure with small grains (Figure 1a). The cross-section image (Figure 1d) reveals a ductile behavior to fracture for the films. Conversely, the stress state changed significantly. DCMS-deposited films were mostly characterized by tensile stresses, while films obtained via HiPIMS showed compressive stresses for thicknesses below 800 nm. The stresses were quite low, in the order of few GPa in absolute value.

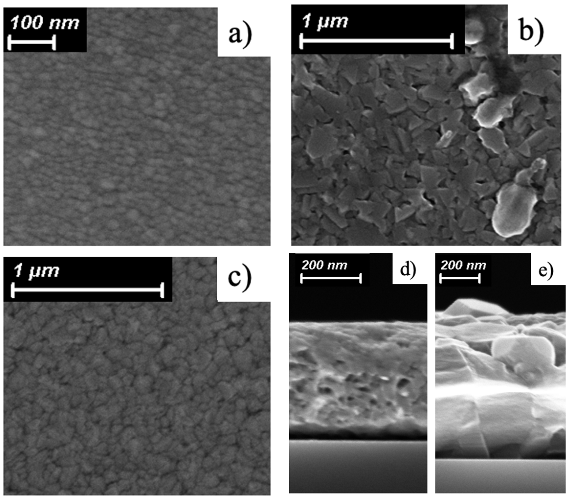

Figure 1: SEM planar images of a) 400 nm HiPIMS sputtered Cu, b) 100 nm DCMS sputtered Al , c) 247 nm biased HiPIMS sputtered Al. SEM cross images of d) 400 nm HiPIMS sputtered Cu e) 800 nm DCMS sputtered Al.

Considering Al, different film morphologies were obtained depending on the deposition regime. In DCMS, the microstructure changed with film thickness, transitioning from small and dense grains (Figure 1b) to larger and coarser ones (Figure 1e). Moreover, the formation of some protrusions (hillocks) was observed (Figure 1b). In the high energy case of HiPIMS, films were characterized by a compact microstructure with small and fine grains even at high thicknesses. Moreover, the formation of hillocks was reduced. Further increasing the species energy and flux by applying a bias had the effect of completely suppressing the formation of protrusions (Figure 1c)). Concerning the stress state, DCMS deposited films showed compression-tensile-compression transitions with increasing thickness, with apparent stress relaxation at higher thicknesses. A similar behavior was also shown by the films deposited with biased HiPIMS. Conversely, HiPIMS-sputtered samples shifted from an initial tensile stress to a compressive one, with no apparent stress relaxation.

3.2. C nanostructured films

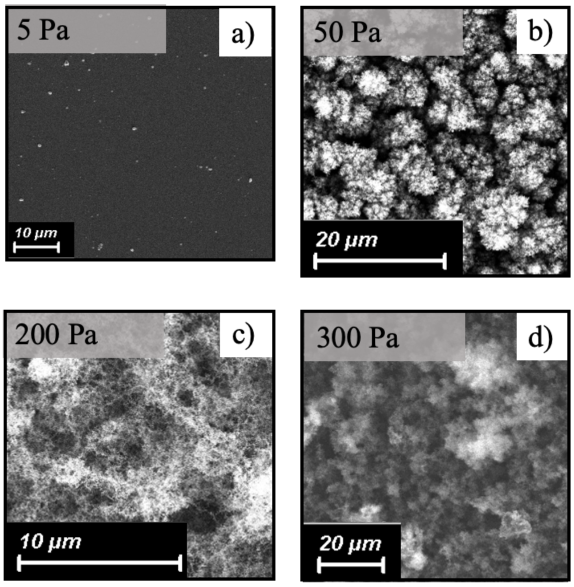

Figure 2: SEM planar view of a) Compact Carbon (CC) deposited via ns-PLD at 5 Pa , b) Tree-like Carbon foam (CT) deposited via ns-PLD at 50 Pa, c) Sponge-like Carbon foam (CF) deposited via ns-PLD at 200 Pa, d) Carbon foam deposited via fs-PLD at 300 Pa.

Carbon nanostrucutred films have been deposited in both $ns-$ and $fs-$ regimes. Concerning ns-PLD, depositions were performed using a circular pyrolitic graphite target, ablated by a laser with a wavelength of 532 nm and measured energy of 500 mJ. Three types of nanostructured Carbon films were produced, varying the Ar pressure in the chamber. At 5 Pa a compact Carbon (CC) film is obtained (Figure 2a), with a density close to the bulk one (i.e., 2000 mg/cm^3). At an intermediate pressure of 50 Pa, a tree-like foam (CT) is produced (Figure 2b), with a density of 26 mg/cm^3. Further increasing the pressure up to 200 Pa, a typical sponge-like morphology (Figure 2c) characterizes the foam (CF), which has a density of 6 mg/cm^3. Concerning fs-PLD, the pyrolytic graphite target is ablated by a laser with a wavelength of 800 nm and a measured energy on target of 3.8 J. One reference sample has been deposited at an Ar pressure of 300 Pa. It is worth mentioning that, despite the higher pressure, the fs-deposited foam is denser than the CF realized in ns-PLD. Indeed the density resulted 7.5 mg/cm^3. Additionally, the surface morphology (Figure 2d)) is more irregular, with the presence of large aggregates. These features are typical of fs-PLD due to the larger aggregates directly ablated form the target.

4. Fishing procedure to realize free-standing films for SL and DLTs

Part of the thesis was devoted to develop a fishing procedure to realize free-standing films. A water solution containing a certain percentage of soap was spin-coated on Si and glass substrates for 1 min. The procedure was repeated one more time to ensure good uniformity. Cu and Al films were deposited according to the parameters described in Section 3. To separate the film from the substrate, the sample was gradually immersed in water. Detachment began when water could penetrate at the interface between film and substrate, gradually lifting off the film. Thanks to its large surface-to-volume ratio, the film could float on water under the action of surface tension. Finally, it was fished with a perforated target holder, obtaining free-standing film in correspondence with the holes. These films could be exploited either as SL or as solid layer in DLT.

Aiming at the realization of the complete DLT, a preliminary investigation of the mutual compatibility between fished free-standing films and C-foams was performed. C-foams were deposited via fs-PLD according to the parameters reported in Section 3.

SEM and Stoney analyses were performed to investigate the effect of the sacrificial layer on films morphology and stresses. Compared to the findings reported in Section 3, the results indicated that the sacrificial layer has no appreciable effects on the stress state nor on the overall morphology. The optimal concentration of soap in the spin-coating solution and the effectiveness of the fishing procedure depends on the combination of film material, substrate, and deposition technique. Cu resulted to be a challenging material. Al samples showed good results when deposited on Si substrates via DCMS and HiPIMS, providing flexibility on the deposition conditions. A spin-coating solution containing 30% of soap (by weight of solvent) was necessary to detach the film. Large-area free-standing films have been obtained with thicknesses ranging from 50 $nm - 800$ nm (Figure 3a).

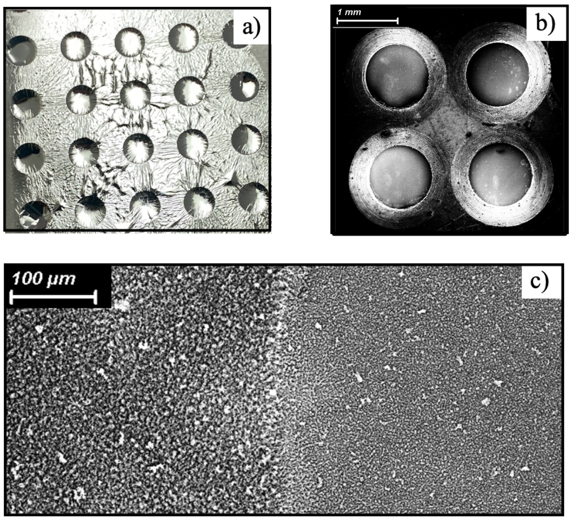

Figure 3: a) Picture of the 50 nm Al free-standing film deposited via HiPIMS, b) SEM planar view of a set of 200 nm Cu free-standing film deposited via DCMS, c) SEM planar view showing the border between the solid holder and the 100 nm Al free-standing film: aggregates on the free-standing films appear to be smaller.

In the case of biased HiPIMS, the depositing species are probably energetic enough to sputter the soap layer and stick to the substrate. Therefore, no detachment of the film occurred. Concerning Cu, it resulted to be a more challenging material. Due to its poor adhesion to Si, glass substrates had to be employed. Promising results were obtained only for Cu deposited via DCMS. In this case, free-standing films ranging from 200 $nm - 2$ μm (Figure 3b) were obtained exploiting a soap concentration of 6% while decreasing the thickness down to 50 nm the 12% was necessary. Cu films deposited via HiPIMS crumbled as soon as they were put in contact with water. To address the feasibility of realizing DLTs, Carbon foams were deposited via fs-PLD on a 200 nm Cu free-standing film, a 100 nm Al film, and on a 50 nm Al film. All free-standing films were covered with good uniformity by the foam. Though, from the SEM image reported in Figure 3c) it is possible to infer a reduction of the amount of deposited material. With respect to the hole-filling technique, the fishing procedure is faster, and enables the realization of larger area free-standing films and the obtainment of much lower thicknesses.

5. C foams for ICF experiments

The other part of the thesis work aimed at verifying the feasibility of producing Carbon nanofoam targets for an experimental campaign that would have been conducted at the ABC laser facility at ENEA Frascati. It was devoted to an explorative investigation of the behavior of C nanofoams irradiated with a laser in ICF-relevant conditions (i.e., ns pulses and intensities $I \geq 10^{14}$ W/cm^2). Simulations [5] demonstrated that in this regime of interaction foam thicknesses of hundreds of μm, corresponding to mass-thicknesses up to thousands of mg/m^2 (when densities of tens of mg/cm^3 are considered), are needed to enable a high laser energy absorption efficiency and conversion in implosion energy (i.e., ablation loading). Foams with these features have never been deposited before at NanoLab. Therefore, the possibility of producing such samples and the mutual compatibility with different kinds of substrate was a critical aspect that had to be investigated for the first time. Additionally, foams with different densities have been deposited as it could have been of interest to investigate the effect of the foam nano/microstructure on the interaction.

Carbon nanostructured films were deposited according to the parameters reported in Section 3. CF, CT and CC samples were realized over a wide range of mass-thicknesses, from ~ 130 mg/m^2 up to ~ 1600 mg/m^2. As substrates, thin (100 nm thick) plastic (CH) free-standing films, thin (1 μm) Al free-standing films, and bulk Al disks were employed. Thin foils are of interest to realize targets suitable for plasma expansion studies via optical diagnostics. They were already mounted on suitable target holders during deposition. The bulk substrate can be exploited to assess the ablation loading by measuring the crater's volume formed as a consequence of shock wave propagation.

As far as CF is concerned, all substrates could endure the deposition, even the one leading to the highest mass-thickness of 1600 mg/m^2 (Figure 4a). Additionally, the resulting film showed good uniformity. The same considerations hold true for CT on Al foils and bulk disks, while plastic foils have not been tested. CC gave rise to some issues. The thin plastic foils were damaged even in the case of the lower mass-thickness samples. Al thin films showed good results as all of them could endure the depositions over the whole range of investigated mass-thickness, even though some of them resulted wrinkled. Additionally, some of the CC films deposited on the bulk Al disk were delaminated. Since compact films have been deposited at low pressure, these issues may be attributed to the high energy of the depositing species that can induce non-negligible stress in the films and, consequently, on the substrate.

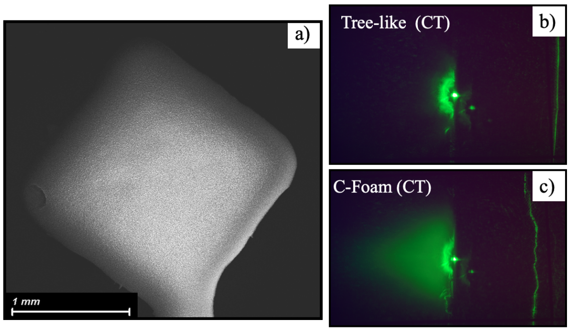

Figure 4: a) Thin (100 nm) plastic substrate covered with CF of 1600 mg/m^2 and optical images of plasma expansion from b) CT 1600 mg/m^2, and c) CC 1600 mg/m^2.

Some of the produced targets were exploited in the experimental campaign. In particular, mass thicknesses in the order of 400 mg/m^2, 800 mg/m^2 and 1600 mg/m^2 have been considered. These targets have been irradiated with a laser having an intensity of 10^{14} W/cm^2, an average energy of 40 J, a pulse duration of 3 ns (FWHM) and a spot diameter of 100 μm. The critical mass density corresponding to a wavelength of 1054 nm is 3.3 mg/cm^3, thus all targets are over-critical. The resulting plasma is probed by a variety of optical diagnostics and radiation detectors. Preliminary qualitative results showed that all the nanostructured Carbon films were transparent to the laser. Nevertheless, substantial differences in the outputs of the several employed diagnostics could be noted when comparing different C-foam samples (see Figure 4b) and c)), suggesting that morphology and density play a crucial role in the interaction.

6. Conclusions

The thesis was focused on the production of nanostructured targets with controlled and tunable properties according to the laser parameters and applications.
In the field of laser-driven ion sources, ultra-high intensity lasers ($I \geq 10^{18}$ W/cm^2) delivering $\sim J$ energies in fs pulses are generally employed, and thin targets are required. A fishing procedure was developed to realize metallic free-standing films via Magnetron Sputtering exploiting a sacrificial layer of soap. By choosing the proper combination of film material, substrate, and deposition conditions, thin large area free-standing films with controlled properties over wide range of thicknesses (from 50 nm up to 2 μm) can be obtained. These films can be exploited either in a SL and DLTs concept. Concerning ICF, the feasibility of producing Carbon foam targets with characteristics compatible with the employed laser parameters ($I \geq 10^{14}$ W/cm^2), energy of tens of J and ns pulses) has been assessed. Carbon foams having mass-thicknesses up to 1600 mg/m^2 have been successfully deposited over different substrates ranging from thin films up to bulk materials.

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