Antireflective coatings obtained by laser-plasma methods for high power laser optical compounds

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In this paper we report on the antireflective coatings with low roughness for high power laser optics obtaining and characterization. The antireflection coatings consist in alternative dielectric oxides layers with low and high refractive index. HfO_2/SiO_2, Al_2O_3/SiO_2, Ta_2O_5/SiO_2, HfO_2/Al_2O_3, Ta_2O_5/Al_2O_3 heterostructures on quartz substrates heated up to 700°C were produced by pulsed laser deposition (PLD) and by radio-frequency plasma assisted pulsed laser deposition (RF-PLD) techniques, in a controllable oxygen atmosphere. The as received structures were investigated by Atomic Force Microscopy (AFM), X-Ray Diffraction (XRD), Secondary Ion Mass Spectrometry (SIMS), Spectro Ellipsometry (SE).

Nowadays, the interest for high power lasers is continuously increasing. In the past, development of such powerful laser systems was limited by the lack of resistant laser materials; since the development of chirped pulse amplification this issue has been resolved for short pulse lengths. Rapid development has taken place in this field, with applications in high-energy-density physics and laser-ignited nuclear fusion. It is expected that at the end of this decade that, for example, the ultrashort lasers power to be increased up to 100 PW.

To fulfil this goal it is critical to develop high-performance laser optics that exhibits elevated laser-induced damage threshold without sacrificing spectral or phase performance. Although the optics can be made larger to lower the power density, this approach inevitably increases both the size and cost of the system.

An important issue arises when high power lasers are involved in the laser-material interaction process. The light "contamination" (prepulse) which arrives before the main pulse from sources such as amplified spontaneous emission (ASE) or higher order dispersion is also focused to high intensities. This prepulse has to be removed using optical devices, because might cause pre-plasma formation, which can significantly change the pulse density and profile, or even destroy targets before the main pulse arrives (see figure1).

The ratio between the main pulse and the prepulse is known as the laser contrast.

An important phenomenon which appears when a high power beam interacts with a (dielectric) material is the generation of plasma (very high densities of the electrons close to solid density - 10^{24} cm^{-3}). At these densities the electrons can follow the field frequency, reflecting the wave. This plasma behaves like a mirror for the electromagnetic radiation. The effect is named plasma mirrors.

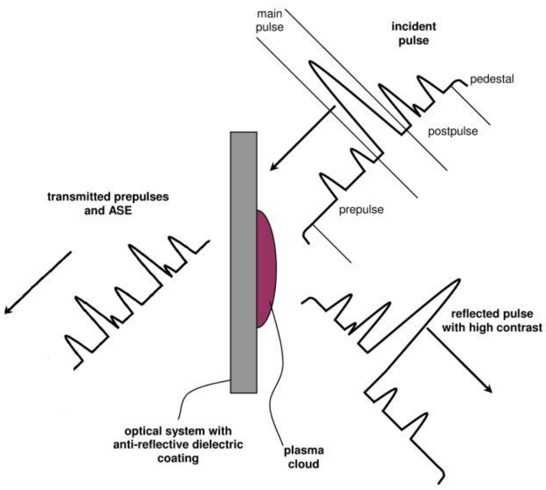

Figure 1. Optical system with anti-reflective dielectric coating

When ultra-short lasers with a specific pulse shape interact with the material, the plasma mirror [1] is generated when the laser pulse is focused such that the main pulse is above the damage threshold of the plasma mirror while the prepulse is below.

The effect will have as result the reflection of the main pulse. Since the laser pulse duration is very short (femtoseconds), the plasma has no time to expand and thus the plasma mirror has the same wave front as the original optic.

Different types of mirrors (i.e. bimorph mirror, off-axis parabolic mirror) were used at the most important facilities in the world, but in the end plasma mirrors were proved to be the most convenient solution to improve the contrast ratio. The use of plasma mirror with antireflection coatings was demonstrated to be an effective solution for applications related to pulse “cleaning”. Indeed, for nanoseconds, picoseconds and femtoseconds pulse duration, specific approaches were used for elimination of prepulse, or amplified spontaneous emission, etc. For example a pair of plasma mirrors reduced the laser prepulse [2], as shown on the picture (Fig. 2).

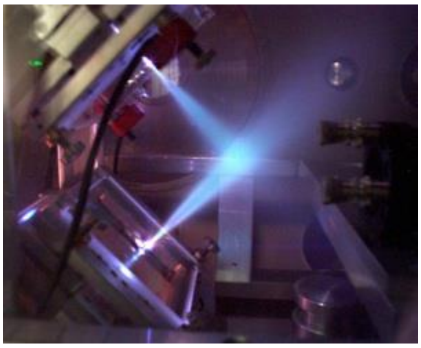

Figure 2. Plasma mirrors

As it was mentioned before, a solution to remove the prepulse is using the use of devices with antireflection coatings. Generally, these coatings are dielectric materials that must have the highest purity to minimize absorption.

These dielectric materials were deposited as heterostructures with different refractive indices by pulsed laser deposition (PLD) and by radio-frequency plasma assisted pulsed laser deposition (RF-PLD).

A reliable and inexpensive method for obtaining thin films of simple or complex compounds is Pulsed Laser Deposition, also called laser ablation. Pulsed laser deposition method involves the interaction of the beam from a laser source with a target material, producing plasma through which it is carried on a substrate, as a thin film. This method has several advantages: a) the deposition chamber is a "clean" reactor as the energy source (laser) is outside the reaction chamber, b) deposition process can be easily controlled because the processes involved are strongly influenced by laser parameters (wavelength, laser fluence, laser spot area, laser pulse duration, repetition rate, etc.), and this control is done from outside the reaction chamber, c) the thickness of the film can be controlled by the number of pulses that irradiate the material, d) the transfer of the target material to the substrate is stoichiometric, f) can produce new materials or in metastable states, that are impossible be achieved by other techniques.

In an alternative ablation process, by irradiating two or more targets, results in complex materials as thin films or heterostructures. The deposition can take place in an inert atmosphere or in the presence of a reactive gas.

Also, a hybrid deposition technique is adopted; this combined the advantages of PLD in an ambient gas (O_2) with the enhanced reactivity associated to a beam of excited and ionized atoms and molecules produced by a radio-frequency (RF) discharge in oxygen and impinging on the substrate. This allows better oxygen incorporation in the layers.

The experimental set-up consists in a laser, a reactive chamber, a pumping system, a heater acting as substrate holder and a rotation-translation system of the target, mass flow controllers for gas admission. A RF discharge generator working at 13.56 MHz and a maximum power of 1000 W is added to the classic PLD system for RF-PLD set-up. For laser ablation experiments different wavelengths where used: λ=1064, 532, 355, 266 nm supplied by a Nd:YAG laser or λ=193 nm from an excimer (ArF) laser.

The combination of the dielectric materials with different refractive indices used as thin films and/or heterostructures with antireflection properties were studied.

In order to obtain high reflectivity for a specific wavelength, the antireflection coatings were formed from alternative oxides layers with low (silica - 1.46 or alumina - 1.77) and high refractive index (hafnia – 2 or tantalalum pentoxide – 2.27). Alternative layers of HfO_2/SiO_2, Al_2O_3/SiO_2, Ta_2O_5/SiO_2, HfO_2/Al_2O_3, Ta_2O_5/Al_2O_3 on quartz substrates heated up to 700°C were produced by PLD and RF-PLD in a controllable oxygen atmosphere; different powers of the RF discharge have been used. The experimental parameters for obtaining of each layer and of layers combination were established after their careful characterization by specific techniques.

The dielectric layers were investigated from morphological, structural, compositional point of view: i) Atomic Force Microscopy (AFM) shows the topography of films, providing information about the surface roughness, features size and aspect; ii) X-Ray Diffraction (XRD) determined the structure, the preferential growth and grains size; iii) Secondary Ion Mass Spectrometry (SIMS) revealed the films composition and distribution of elements in the deposited layer or heterostructures; iv) Ellipsometry investigation determined the refractive index, offering also details about layers thickness and roughness.

[1] C. THAURY et al., Nature Physics, 3, 424-429 (2007)

[2] D.M. Gold et al, Opt. Lett.19, 2006 (1994)