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TITLE INVESTIGATION OF METAL COATINGS FOR THE FREE ELECTRON LASER

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Investigation of Metal Coatings for the Free Electron Laser*

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We are investigating the deposition and characteristics of metal coatings for use in environments such as the Free Electron Laser where the radiation resistance of metal coatings could prove to be of great benefit. We have concentrated our initial efforts on silver laminate coatings due to the high reflectance of silver at 1 micron wavelength. Our initial laminate coatings have utilized thin layers of titanium oxide to break up the columnar structure of the silver during electron-beam deposition on fused silica substrates. Our initial results on equal coating thickness samples indicate an improvement in damage threshold that ranges from 1.07 to 1.71 at 351 nm.

Key Words: metal coatings; silver coatings; coating structure; laminate coatings; laser damage.

1. Introduction

The Free Electron Laser is a hostile environment for the optical coatings typically used in a laser cavity. Radiation, such as x-rays, gamma rays, neutrons, and scattered electrons are present in the laser cavity due to collisions of the high energy electrons from the accelerator with magnets, walls, etc. Dielectric materials used in multilayer reflectors are sensitive to this radiation environment and the performance of a multilayer dielectric coating as well as its laser damage threshold can be quickly degraded in response to these radiations.

Previous work at Los Alamos on aluminum films indicated that the physical properties of aluminum films could be improved by interrupting the growth of columns in the aluminum films with pulses of oxygen forming thin oxide layers periodically during the aluminum deposition [1,2,3,4]. Similar work was conducted at Los Alamos with tantalum films and pulsed Acetylene gas [5]. A TEM photomicrograph of one of these aluminum laminate structures is shown in figure 1. The yield strength of these aluminum films increased 20 fold from 20 MPa to 400 MPa. In addition, unbacked cylindrical aluminum films only 2000 Angstroms thick were deposited for the Inertial Confinement Fusion Program [6].

This previous work on aluminum films encouraged us to look

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for a similar technique to use with silver films. One difficulty in extending this technique to silver is that the silver film does not readily oxidize in the presence of oxygen. Thus, we were led to depositing a thin layer of another material to interrupt the structure of the silver films. The material we chose to use in this laminate structure is titanium oxide. A thin layer (50 Angstroms) of non-stoichiometric titanium oxide was deposited periodically during the deposition of silver films on fused silica substrates to investigate its effectiveness in improving laser damage threshold (see figure 2).

2. Film Analysis

An Auger depth profile of one of our first silver laminate films is shown in figure 3. The titanium oxide layers are well defined in this structure, though the oxide layers are only 50 to 85 Angstroms thick. A SEM photomicrograph of the cross-section through this laminate structure lacked sufficient resolution to see structure effects in these films. TFM photos have not yet been obtained. SEM photos of the surfaces of silver laminate films and typical silver films indicate that the laminate surfaces are smoother (see figures 4 and 5).

3. Film Fabrication

We prepared our silver films using electron beam deposition in a high vacuum chamber. The deposition rate of the silver layers was relatively high at 25 Angstroms/sec. Thin layers of titanium metal were evaporated periodically and this thin layer was immediately oxidized by introducing a pulse of oxygen into the chamber. All of the samples to be reported here were deposited on highly polished fused silica substrates, 5.08 cm in diameter and 0.955 cm thick.

Two samples each of 4 coating designs were deposited for this study. The first coating design is a 2000 Angstrom thick silver film reference deposited on a 100 Angstrom layer of chromium. The second design is a 4 layer laminate beginning with a 50 Angstrom titanium oxide layer, followed by a 1500 Angstrom silver layer. This layer pair is repeated to complete the 4 layers. The third coating design is similar to the second, except that there are 12 layers, or 6 pair of titanium oxide and silver layers, instead of 2 pair. The fourth coating design is a 9200 Angstrom thick silver reference film deposited on a 100 Angstrom chromium binder layer. This fourth design is a silver reference of equal thickness to the 6 pair laminate of the third design. Comparison of equal thickness samples is required due to the thermal insulation of the substrate.

4. Surface Roughness Measurements

Surface roughness measurements were performed on these silver

samples using a Wyco optical heterodyne profilometer. This non-contact method of determining the microroughness of an optical surface will not affect the subsequent laser damage threshold measurements. The results of these measurements on our 8 samples are given in table 1. The thin samples appear to be somewhat rougher than the thicker samples, however, all of the samples are less than 9 Angstroms rms along measurement lengths of 1.2 mm.

Table 1. Silver film surface roughness measurements

Sample #	Design	RMS Roughness(A)
1	100A Cr/2000A Ag	8.4
2	100A Cr/2000A Ag	7.9
3	[50A TiOx/1500A Ag]*2	7.5
4	[50A TiOx/1500A Ag]*2	7.7
5	[50A TiOx/1500A Ag]*6	6.3
6	[50A TiOx/1500A Ag]*6	5.1
7	100A Cr/9200A Ag	5.9
8	100A Cr/9200A Ag	6.7

5. Reflectance Measurements at 351 nm

Reflectance measurements were made on the 8 samples using a W-V attachment on a spectrophotometer from 300 nm to 500 nm. These spectrophotometer measurements are shown in figures 6 to 9. These spectral traces show the emergence of the well known surface plasmon resonance in silver (at 345 nm), particularly in the thicker samples. Since the wavelength of the laser performing the damage tests conducted on these samples is 351 nm, the reflectance values at this wavelength have been summarized in table 2. The silver film absorption (1-R) varies between 18% and 36% in these samples due to the variation in the surface plasmon coupling with film thickness. Surface plasmon coupling variation with surface roughness is well known, but the variation in coupling with thickness is not explainable on this basis.

Table 2. Silver film reflectance measurements at 351nm

Sample #	Design	Reflectance(%)
1	100A Cr/2000A Ag	77.8
2	100A Cr/2000A Ag	77.8
3	[50A TiOx/1500A Ag]*2	76.0
4	[50A TiOx/1500A Ag]*2	67.0
5	[50A TiOx/1500A Ag]*6	68.6
6	[50A TiOx/1500A Ag]*6	72.5
7	100A Cr/9200A Ag	68.2
8	100A Cr/9200A Ag	64.2

6. Laser Damage Threshold Measurements at 351 nm.

Laser damage tests of these samples were conducted at Los Alamos in Steve Foltyn's XeF (351 nm) laser damage laboratory by John Jolin. The absolute laser damage thresholds determined in these tests are not important in our investigation. Thus, the high absorption in the coatings and the substrate being a thermal insulator do not alter the conclusions of our tests. The relative damage performance of the laminate coatings when compared to the reference silver coatings of the same thickness illustrates any improvement due to the laminate design. The results of these laser damage tests are given in table 3.

Table 3. Silver film laser damage thresholds at 351nm

Sample #	Design	Laser Damage Threshold(mJ/cm ²)
1	100Å Cr/2000Å Ag	50
2	100Å Cr/2000Å Ag	50
3	[50Å TiO ₂ /1500Å Ag]*3	80
4	[50Å TiO ₂ /1500Å Ag]*2	70
5	[50Å TiO ₂ /1500Å Ag]*6	150
6	[50Å TiO ₂ /1500Å Ag]*6	240
7	100Å Cr/9200Å Ag	140
8	100Å Cr/9200Å Ag	140

The effect of thin film thickness is clearly evident when one compares the damage thresholds of the last 2 samples to the first 2 samples. These samples are identical silver films except for thickness. The first two samples have a total metal film thickness of 2100 Angstroms each and the last 2 samples have 9300 Angstroms each. This increase of 4.5 times in coating thickness results in a factor of 3 increase in laser damage threshold of the silver reference films.

The most significant results of these laser damage tests are seen in comparing the thresholds measured for the 6 pair laminate structures (samples 3&6) and the equal thickness reference silver films (samples 7&8). The two thick reference samples each measured 140 mJ/cm², whereas the laminate films measured 150 mJ/cm² and 240 mJ/cm² for samples 5 and 6 respectively. Thus, there is an improvement in the damage threshold of a factor of 1.07 to 1.71 for the laminates over the silver reference films. We cannot account for the large discrepancy between the damage thresholds of samples 5 and 6, except to note that these coatings were deposited in 2 separate coating runs using the same prescription. Perhaps further investigation of these samples will reveal some difference in the coatings.

7. Conclusions

We have concluded from this preliminary investigation of silver films that laminate structures such as the ones used in this study may prove to be useful in increasing the laser damage thresholds of metal films. This technique could also be extended to dielectric thin films for use in multilayer coatings.

The authors would like to express their appreciation to Steve Foltyn and John Jolin of Los Alamos for performing the laser damage measurements quoted in this paper. We would also like to acknowledge Glen Lindholm and Kathleen Padgett of Los Alamos for performing the spectrophotometer reflectance measurements.

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Figure 1. TEM photomicrograph of an aluminum/aluminum oxide laminate film in cross-section. Note the thin oxide layers separating the thick aluminum layers.

80 nm

Figure 2. A schematic view of a silver/titanium oxide laminate film with 50 Angstrom titanium oxide layers and 1500 Angstrom silver layers.

Figure 3. Auger depth profile of a silver/titanium oxide laminate film.

Figure 4. SEM photo of the surface of a typical silver film.

Ag/Ti Laminate
Ag Surface

000004 25KV X25.0K 1.20um

Figure 5. SEM photo of the surface of a silver/titanium oxide film.

Figure 6. Spectrophotometer reflectance traces of samples 1 & 2 with a reference curve calculated from published optical constants of silver.

Figure 7. Spectrophotometer reflectance traces of samples 3 & 4 with

Figure 8. Spectrophotometer reflectance traces of samples 5 and 6 with calculated silver curve.

Figure 9. Spectrophotometer reflectance traces of samples 7 and 8 with