Reducing the Stress of Hafnia/Silica Multilayers with Ion Assisted Deposition for Use in High-Power Diffraction Gratings

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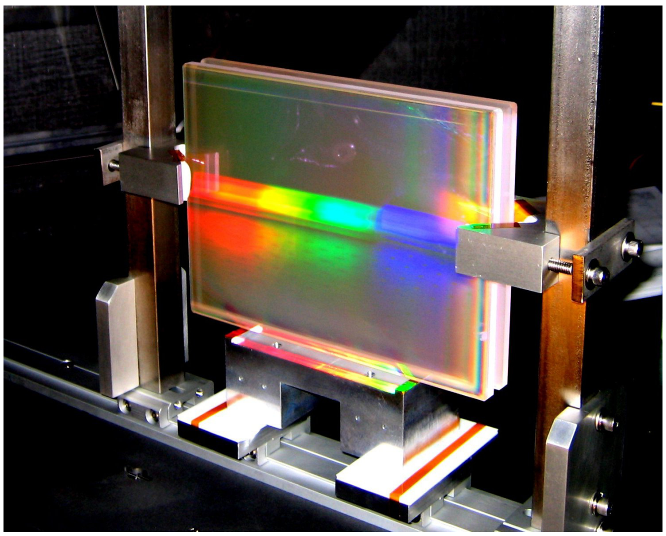
Key Words: Residual Film Stress; Ion Assisted Deposition; Laser Damage Threshold; Electron Beam Deposition



Abstract

Multilayer dielectric optical films are used to achieve high laser-induced damage threshold coatings for mirrors in high energy lasers. These coatings, usually made without ion-assisted deposition (IAD), can have low stress in a typical laboratory atmosphere on any substrate material. However, some applications, such as the generation of time-compressed pulses require a vacuum environment for propagation of the laser pulse. In these applications, films on low thermal expansion substrates develop excessive tensile stress. Ion-assist using an RF ion source is used to increase the compressive stress in the multilayers to achieve a low stress, high laser damage threshold multilayer coating on a silica substrate in vacuum. These coatings are suitable for use in an all-dielectric diffraction grating (MLD).

Introduction

Multilayer diffraction gratings (MLD) are used to temporally compress the pulse in high energy lasers from 1 ns to less than 1 ps [1]. These gratings are used with large beams (41cm x 41cm) at 1054 nm and at an angle-of-incidence of 65-75º. This high incidence angle requires very large gratings; as large as 1.5 meters in length by 0.5 meters in width. Limitations in the size of fabricated gratings force the designers to use segmented grating optics consisting of two or three segments to make up a large grating [2]. Additionally, the time compressed pulse will have an electric field component that will exceed the breakdown of air, so the gratings must be used in a vacuum environment. These conditions place strict requirements on the allowed stress of the coatings used for MLD gratings.

Coating stress has two detrimental effects on thin films: It causes the substrates to bend, and, if sufficiently tensile, it can cause coatings to fracture. The shape formed by stress of a uniform isotropic coating on a homogeneous “thin” substrate will be a sphere. At an oblique angle, the diffracted wavefront from this shape will be composed of power (focus) and astigmatism. These Seidel aberrations can usually be removed by a deformable mirror placed before the grating. However, in a tiled structure, the combination of several gratings, as in Figure 1, can produce a much more complex wavefront. This wavefront is not easily corrected due to the high frequency components occurring near the mirror boundaries.

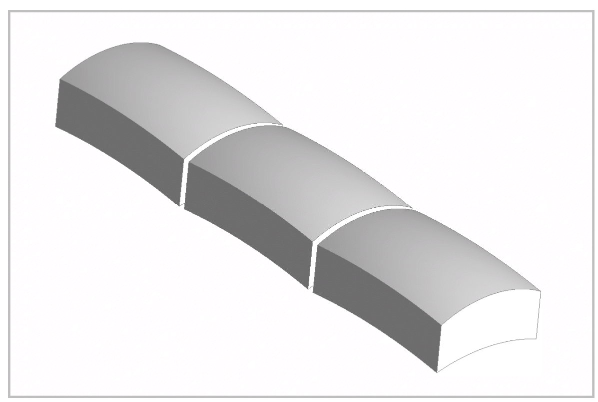


Figure 1. *Three stressed grating substrates tiled together to form one large grating. The coatings in this diagram are shown in compressive stress. The tiled component produces a complicated wavefront when the individual optics are under uniform stress as shown here.*

In tension, high stress will cause both wavefront deformation and coating and substrate fracture failure. This failure, often called “crazing”, is caused when the tensile stress exceeds the fracture strength of a substrate or coating flaw. Typically, the crack continues into the substrate to a depth many times the thickness of the film. Figure 2 is a scanning electron micrograph (SEM) of a

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craze line in a substrate. The final depth of the craze was about 30 microns, more than 5 times the thickness of the thin film! The shape of the fracture is responsible for the changing brightness of the craze lines when they are viewed in different lighting. Note that the width of the craze fracture in this picture is only a few hundred nanometers. Craze lines will always form first at defects in the substrates. These could be subsurface flaws which remain from an earlier grinding operation applied to the surface.

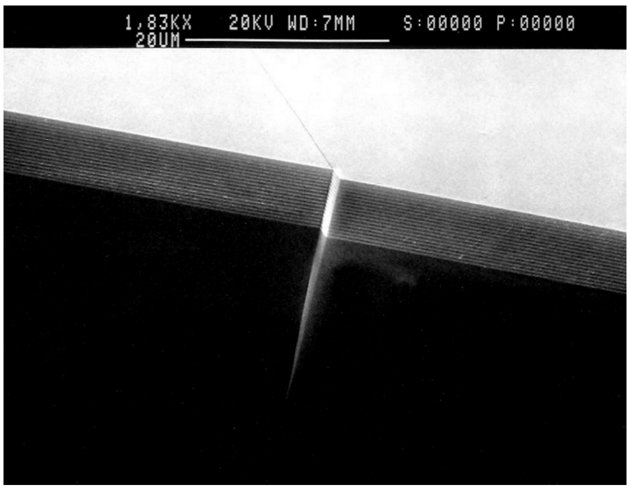

Figure 2. *A SEM of a craze fracture in a substrate. The fracture was formed by over-baking a coated BK7 substrate. A multilayer film of about 6 microns total thickness is seen on top and was the source of stress which formed the fracture. The fracture penetrates into the substrate below more than 30 microns.*

Thin film stress has several origins [3]. The predominant origins are due to thermal mismatch and film structure. Stress in oxide films is dependant on three conditions: temperature and thermal properties of the substrate, water vapor (humidity), and time (see equation 1 below). Previous work [4] found that e-beam deposition could produce low stress films on both low coefficient of thermal expansion (CTE) substrates, such as silica and Zerodur®, and high CTE substrates, such as Schott BK7 glass for optics used in an ambient environment (relative humidity = 45%). In these cases, the intrinsic stress was controlled by varying the oxygen back pressure during deposition of the silica layer. However, in a dry environment low stress coatings could be made only on BK7 substrates; coatings made on a low CTE substrate, such as silica, will always have excessive tensile stress. In these cases the coatings used a hafnium oxide / silicon dioxide layer paired to achieve reflectance requirements and a high laser damage threshold for 1054 nm pulses. The coatings were depositied on substrates held at 200 degrees C. The process conditions for the hafnium dioxide are determined by the requirements for a high damage threshold and do not have a significant influence on stress.

[Equation 1.]

$$\sigma_{total} = \sigma_{thermal\ mismatch} + \sigma_{H2O} + \sigma_{intrinsic(time)}$$

Absorbed water vapor plays a key role in the stress performance of the coating. Water vapor is absorbed into the columnar structure and causes both short and long term changes in the film stress (see Figure 3). Oxide films are especially susceptible to this effect. The dangling bonds of the SiO_2 films attract the polar water vapor water molecules from the atmosphere. In previous work, both spectral performance shift and stress shift were found to be highly dependant on the relative humidity of the air in the test environment [5]. The change takes places rapidly in these multilayers. The stress ceases to change only 5 minutes after changing the atmospheric water vapor level. However, a long-term aging effect in the stress also occurs. This is believed to be due to microscopic changes in the film structure in response to the water-induced stress (figure 4). The water can also be removed chemically by rinsing the coating with a strong acid, such as formic acid. These phenomena have been the cause for tensile failure in a number of films requiring post-processing.

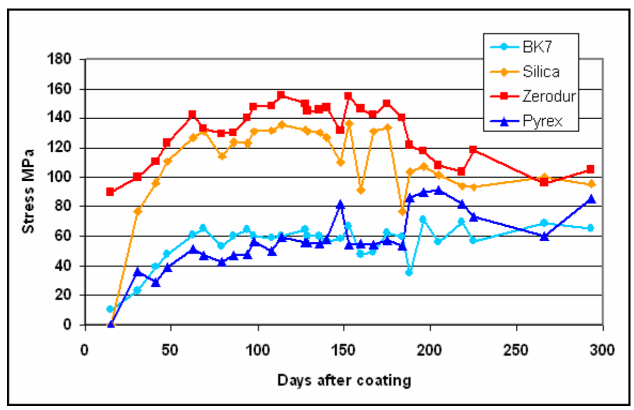

Figure 3. *The effects of aging and substrate type on stress of an electron-beam deposited polarizer coating. All measurements were made in dry Nitrogen. The low CTE substrates Zerodur® and silica drift into tensile stress soon after coating. The higher CTE substrates are "pre-stressed" by coating at elevated temperatures and are always more compressive. Most aging effects occur in the first month after coating.*

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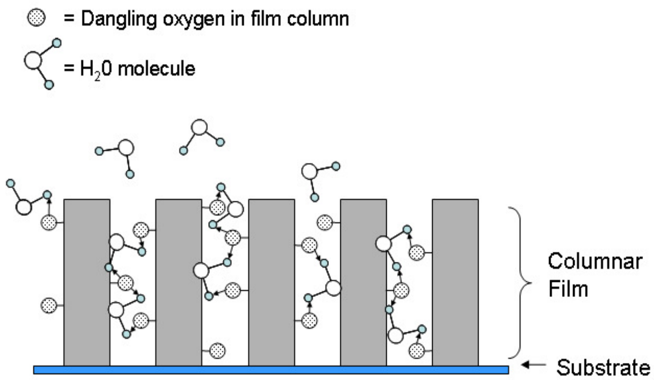


Figure 4. ***Water vapor causes both short and long term changes in the stress of the film.*** *Bipolar water molecules attach to dangling bonds in the columnar structure of the films. Migration of the water molecules in and out of the coating causes short term changes in stress; changes in the columnar structure of the coating due to pressure by the water is believed to cause the long term changes in stress.*

The choice of a low CTE substrate for the grating is preferred since fabrication, measurement, and grating specification requirements would all be best met by a low-expansion substrate. However, conventional e-beam deposition cannot produce low stress films when they are used in a dry atmosphere. Figure 3 shows many features of e-beam deposited film stress on substrates with a different CTE. Generally, the low CTE substrate films will climb into a region of high tensile stress after a short period of time. Depending on the substrate geometry, films with stresses ranging from 10 to 100 MPa will be sufficient to bend an optical substrate out of specification. For grating applications 20 MPa is considered a reasonable upper limit. Additionally, these coatings must operate in a vacuum because as the pulse width decreases, the power density exceeds the breakdown strength of air. The films must be made more compressive for low CTE substrates to be suitable. One method to add compressive stress to an electron-beam deposited oxide film is to use ion-assisted deposition. The distinct advantage of this method is that it is scalable to the larger sized substrates. The questions which must be resolved in this study are: 1) What are the correct conditions to produce a low stress coating in the vacuum environment of the grating chamber, and 2) Can a low stress, ion-assisted coating be made with a high damage threshold for 1054 nm light?

Experiment

A study of the stress and laser damage was conducted on a series of IAD coatings. The coatings were made in a 1.1 meter box coater equipped with a Veeco 12 cm gridded RF ion source. An RF neutralizer is used to produce electrons to allow a charge balance to occur after the energy of the ions is dissipated. The absence of filaments in the ion source may be key, since contamination from filaments has been shown to effect damage thresholds in the past [6]. The coatings were designed and deposited using hafnium oxide and silicon dioxide. The design used 26 layers and was a simple reflector with matching layers for a MLD grating to be used at a range of 62 to 72.5 degrees angle-of-incidence. The design was optimized using a grating simulation program. The last layer is intended to be used as the grating layer, however, none of the films examined at this stage of the study were etched to produce a grating structure.

The 12 cm gridded ion source uses a Molybdenum 3-grid ion optics arrangement. The grids are dished to produce a divergent beam. The three-grid arrangement allows a high beam current at low beam voltages with low erosion of the grids. The discharge is produced by an RF antenna which encloses a quartz discharge chamber. This design introduces a low contamination level of material sputtered from the ion source into the deposited thin film. The source also has high reliability and requires very little maintenance. It is configured to be operated with Oxygen, Argon, or a mix of both gases. The RF neutralizer must be used with Argon gas so there is always a level of this gas in the chamber during deposition. The ion source can be configured to automatically switch operating parameters and gases between layers so the programs for the HfO_2 and the SiO_2 layer can be quite different. Sometimes an intermediate program is required to prevent preferential sputtering of oxygen from one of the layers.

Multiple IAD experiments were done to determine the correct ion source and coating parameters for producing low stress on silica substrates (Figure 5). These experiments were done in two stages. All coatings were a full, 26 layer MLD grating design. Only the parameters on the silica layers were altered. Parameters on Hafnia layers remained constant throughout the entire experiment. All experiments used two substrate types: fused silica, (low CTE) and Corning 0211 (high CTE, similar to BK-7).

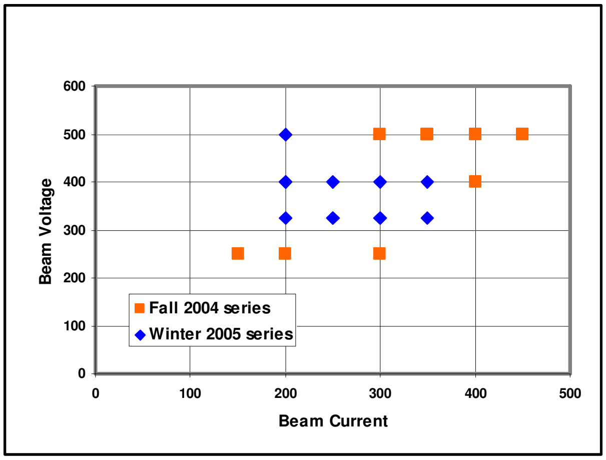


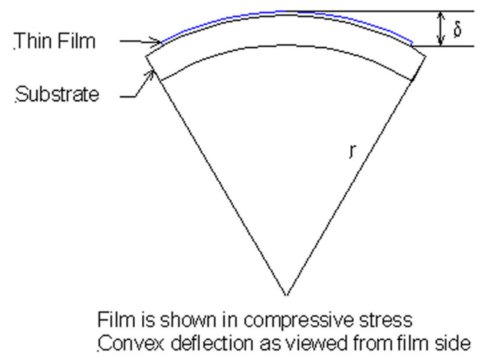
Figure 5. *Ion-assisted deposition experiments were done in two stages. The plot shows the ion source beam voltage and current parameters used in each stage.*

Stage 1 (Fall 2004) experiments consisted of a series of high and low ion source power coatings, in an attempt to bracket the effect of ion source parameters on stress. This initial stage was flawed by inadequate silica substrates. The samples were too thick, and inconsistent substrate flatness produced arbitrary and unreliable measurements. Coatings in Stage 2 (Winter 2005) filled the ion beam power gap left by Stage 1. Some flawed runs from the previous stage were repeated with the new, substantially thinner, silica substrates. Collectively, over 50 coating runs were conducted for this study, 10 of which were Non-IAD runs.

Stress Calculation and Analysis

Stress samples from each run were measured and analyzed at specific time intervals.
Calculating film stress requires the evaluation of several factors, as described previously. Stress analysis in this study focused on controlling and monitoring relative humidity and film age. The radius of each sample is measured on an optical profilometer in both lab ambient and dry nitrogen environments. A phase shift interferometer is used to measure larger samples, in which stress is calculated in terms of sag. Equation 2 in Figure 6 shows a simple conversion of radius to curvature in terms of sag. Once converted, the Stoney equation (equation 3) is used to derive the stress coefficient of film (σ).

Coated samples are measured over a period of 6 months or more. Standard stress measurements are made at 1 week, 2 week, 1 month, 2 month, 3 month and 6 month intervals after coating. It should be noted that stress changes rapidly within the first week. These early measurements can often produce ambiguous data. Surface contamination, sample reversal and ambient humidity variations may also effect stress measurements. In addition to tracking internal stress shift over time, multiple measurements of each sample confirm accurate data, and flag the ambiguous.



EQ. 2

$$\delta = \frac{D^2}{8r}$$

EQ. 3

$$\sigma = \frac{4E_s t_s^2 \delta}{(1-\nu)3D^2 t_f}$$

r = radius of curvature
D = diameter of substrate (or clear aperture)
δ = sag of stressed plate
σ = stress coefficient of film
E_s = Young's modulus of substrate
T_s = substrate thickness
υ = Poisson ratio of substrate
t_f = thickness of film

Figure 6. *Stoney equation for finding the stress of a thin film.*

Laser Damage Testing

Some of the coatings were also deposited onto a 50mm diameter by 10 mm thick substrate for damage testing. Laser damage tests of samples were provided commercially by Spica Inc. using a 1064 nm laser with a pulse width of 3.5 ns. A scanning method was used where by a 1.1 mm beam was scanned over a 1 cm^2 area at a given fluence. After, or during the scan, the substrate is observed for damage indicated by scattered light. The fluence is then increased and the process is repeated. The highest fluence at which there is no observed damage is the "qualified" fluence. The fluence range at which damage occurs but does not grow on subsequent shots is called the "probable" region. The fluence where damage propagates into larger, catastrophic sites is the "fail" threshold. The scanning method closely follows techniques developed for testing optics for the National Ignition Facility (NIF). Generally, an optic operating in the probable region will be usable.

Results

The stress of a multilayer grating coating on fused silica and borosilcate substrates for a range of ion gun powers is given in figure 7. The power is recorded at the ion source; no ion fluences in the substrate plane were recorded in this phase of our study (this will be done prior to scale-up of the method to a larger chamber). The power also refers only to the power used during the deposition of the silica layers. The power during deposition of the hafnia layers was kept constant throughout the study. Since the coatings were made at a low temperature (60 to 90 degrees C), there is much less difference in the response of the two substrate materials. The graph in figure 7 shows two phenomena: 1) a compressive film on silica (or BK7) can be made by using ion assist and, 2) a grating multilayer can be made with near zero stress on both types of substrates. All measurements in the graph were made on films aged 4 weeks or more and measured in a nitrogen atmosphere.

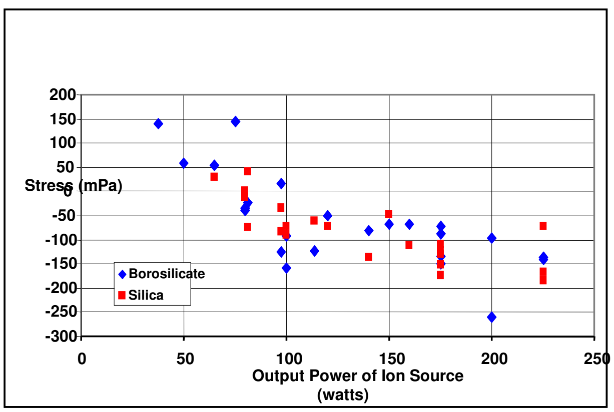


Figure 7: *The stress in coatings versus the ion source power. Sample age is greater than 4 weeks, and all measurements are made in dry Nitrogen.*

Figure 8 shows the time evolution of stress of two samples made at 80 watts ion output power of the ion source. This was a quick experiment to show the repeatability of the best results. Both runs were tested shortly after and then 2, 4, 8, and 12 weeks after coating. The progression in stress is fairly similar, starting with a high compressive stress of 60 to 70 Mpa and relaxing to less than 10 MPa of stress after two months of aging. Future tests will include a large number of samples to verify these results. The damage results for a sampling of the IAD coatings are shown in Figure 9. The damage thresholds of many of the IAD coatings were equivalent or greater than the traditional electron-beam coatings. This data will be supplemented with testing at short pulsewidths and also on grating structures at a later time.

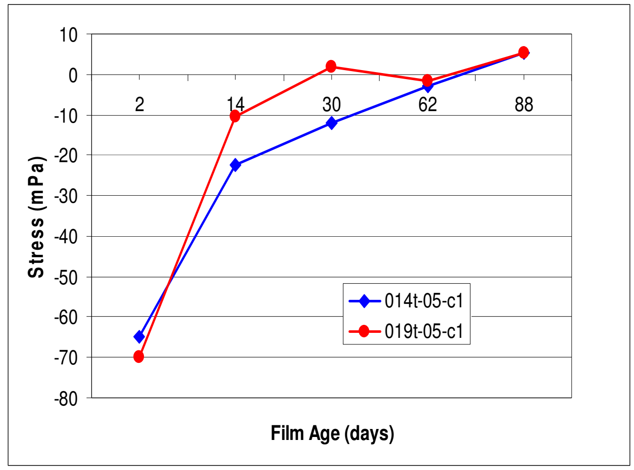


Figure 8: *Stress evolution from two identical coating runs of the best results on silica. Measurements taken in simulated vacuum environment*

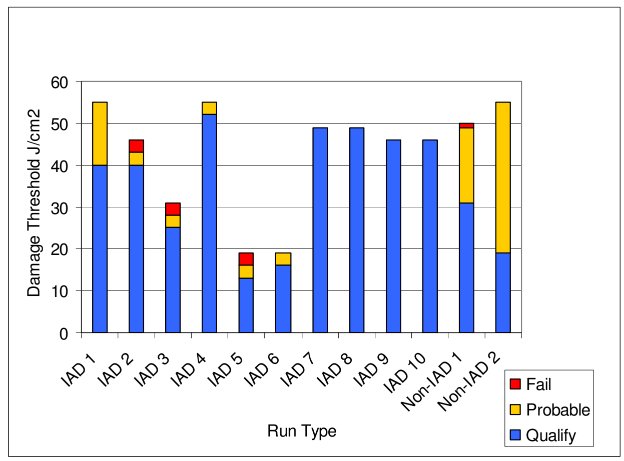


Figure 9: *Damage test results for various IAD and Non-IAD e-beam coated samples. Qualified regions indicate no damage. Damage is initiated but does not grow in the probable regions. Failure indicates that damage is either growing or catastrophic.*

Conclusions

The stress in a multilayer oxide film on a fused silica substrate can be reduced to less than 20 MPa when measured in a dry nitrogen atmosphere by using ion assisted electron- beam deposition. The damage thresholds of the ion assisted coatings were measured with 3.5 ns pulses and were comparable or greater then non-IAD coatings. Future efforts will include testing at short pulsewidths (0.5 to 10 picoseconds) of both coatings and MLD gratings and finally scale-up of the process to large substrates (0.9 x 0.45 meters). Stress of the coating in vacuum should be compared to stress measured in a dry nitrogen environment.

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