

Co-deposition of double layer antireflection coatings and laser induced damage threshold (LIDT) measurements at 1064 nm: Inhomogeneous systems

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Inhomogeneous antireflection coatings were prepared by simultaneous evaporation of high and low index materials during production of the interface region. Rates of evaporation for both materials were monitored by microprocessor controlled quartz crystals. Four designs were prepared, three of them consist of both oxides (Hafnia, Tantalum, Zirconia / Silica) and the other one contains Cerium oxide in combination with Magnesium fluoride. Absorption and damage threshold values are presented.

(Received March 13, 2008; accepted August 14, 2008)

Keywords: Evaporated inhomogeneous coatings, Antireflection coatings, High damage threshold coatings

1. Introduction

The major concern of the high damage thin film researchers is to enhance the damage threshold by using different techniques so that dielectric AR, HR or any other coatings become suitable for high power laser applications. A literature survey regarding inhomogeneous coatings revealed that the research has proceeded in two directions. 1) Theoretical aspects in which various models were developed and damage threshold were calculated [1-4,6,9,10,13,15,17-19]. 2) Experimental aspects in which damage threshold were measured as a function of time domain of laser pulses, impurity diameters, absorption etc. Moreover, automatically controlled deposition methods were developed for reproducibility of the coating designs [5,8,12,14,20]. Due to the continued interest in high damage threshold optical coatings, especially double layer antireflection coatings for high power laser application are of special importance [5,11,21-23]. The basic idea for LIDT enhancement evolved from the influence of interface absorptions between two consecutive layers using co-deposition technique. [7,14]. This paper describes that absorption losses may order of magnitude higher in homogeneous layer systems with abrupt interface than in gradual interface inhomogeneous layers prepared by automatically controlled evaporation process.

It is now more than two decades that the concept of evaporated inhomogeneous thin films was realized [1, 4]. The inhomogeneous layers could not get much attention, because of the development of homogeneous thin dielectric systems. During the last few years, the interest in inhomogeneous coatings has been revived due to the development of automatically controlled evaporation

systems. In such processes the rate of evaporation can precisely be controlled for different materials separately. In order to produce an inhomogeneous layer or system of layers, two materials with different refractive indices are evaporated simultaneously from two sources. The rate of both materials can be varied as a function of time. The inhomogeneity consists of a gradient of the refractive index in the direction z perpendicular to the film surface. This situation is described in figure 1 where n varies continuously as a function of z , i.e., $n=n(z)$.

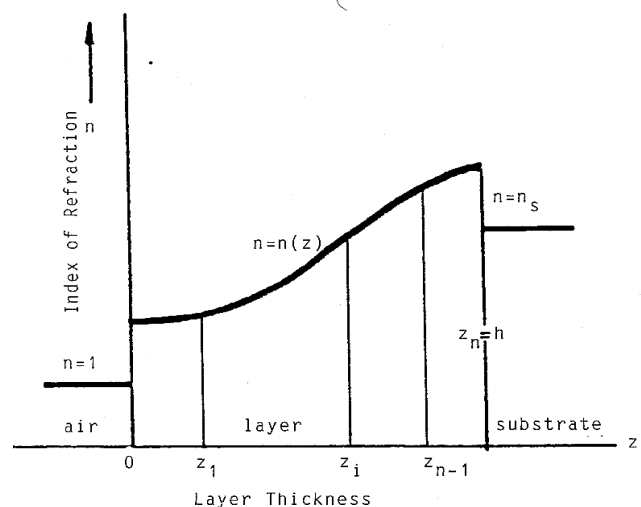


Fig. 1. Refractive index variation profile of an inhomogeneous layer.

Normal dielectric multilayer coatings with abrupt interfaces exhibit heavy absorption losses at film-film interface [14], which may contribute to damage mechanisms. It is now possible to minimize these losses by evaporating both high and low refractive index materials during the interface region so that the system behaves like a single layer without any abruptness. In the first part of these investigations, we have studied homogeneous antireflection systems [24]. Broad angle antireflection coatings were reported in [25]. This paper describes the results of inhomogeneous or gradual interface double layer AR-coatings.

2. Experimental setup

Double layer antireflection systems with inhomogeneous or gradual interfaces were prepared in a Leybold- Heraeus vacuum coating unit. Two crystal monitors were mounted above each E-gun source for monitoring the rate of evaporation. Both materials are evaporated simultaneously from two E-gun sources. A microprocessor (Z-80) was used to control the whole evaporation process. The schematic arrangement is shown in figure 2. The sub-system consist of a frequency counter for the quartz crystal monitors (M), B is the control panel and D/A converts digital to analog for controlling the power supply.

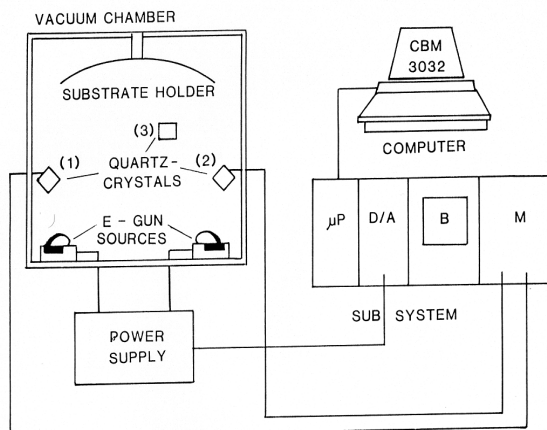


Fig. 2. Schematic of vacuum chamber for the preparation of inhomogeneous

Four AR-2 designs, Hafnia/Silica, Zirconia/Silica, Tantalum/Silica and Cerium oxide/Magnesium fluoride were selected for coatings. Initially, high index material was evaporated at normal rate of evaporation 'as the on set point reaches where the interface begins, both materials start to evaporate.

A mixture of high and low index material is deposited to form a continuous interface. As the deposition time (in our case it is kept constant to 40 seconds) is over, the emission current for the source 2 goes to zero and the low index material is further deposited until the transmission of the AR-coating reaches to a maximum. The rate of evaporation for both materials is registered by the

computer. Each material was calibrated as a function of film thickness vs. frequency shift of both the Quartz crystals separately in nm/Hz.

In preparing inhomogeneous AR-coatings, an arbitrary wavelength shift was noticed due to the co-deposition effect which in turn shifts the reflectance minima. In order to get minimum reflectance at central wavelengths, three samples were prepared with different onset points. The position of the onset was selected arbitrarily. In the same batch, abrupt AR-2 systems were also prepared for the comparison of absorption and damage threshold with co deposited systems. LIDT measurement details have been reported elsewhere [24].

3. Results and discussion

3.1 AR-2 systems with gradual interfaces

From the recorded data of rate of evaporation for different inhomogeneous systems, the index profile can be obtained as a function of physical thickness of the film and time for each set of data. As an example, we present the Hafnia / Silica design. Fig. 3(a) shows the index profile n as a function of time and figure 3(b) gives the refractive index n as a function of physical thickness of the interface.

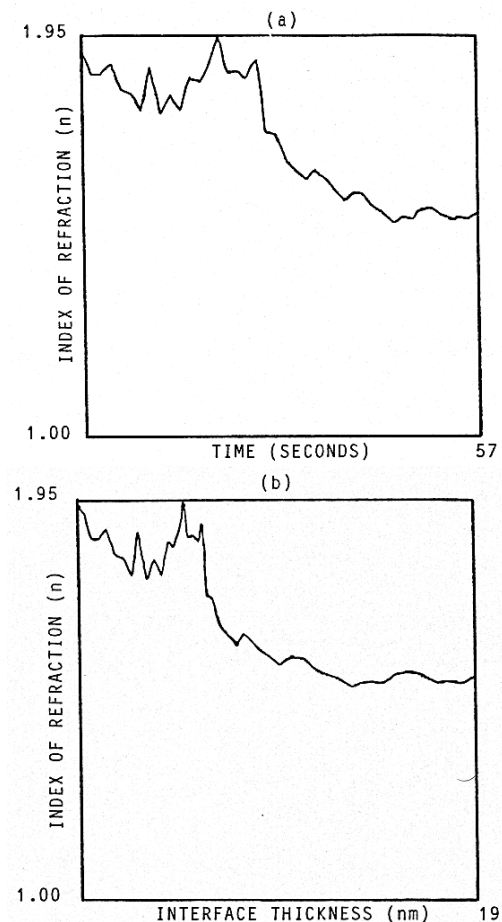


Fig. 3(a) & (b) Refractive index profile of inhomogeneous AR-2 with Hafnia / Silica as a function of time and film thickness.

It is evident from the figure that refractive index varies continuously from high index (1.95) to the low index (1.46) with time. After approximately 20 seconds, a bump in the figure is visible. This is due to the fact that low index material starts to evaporate not exactly at the beginning of the interface but a little bit later. Although the evaporation time was fixed to be 40 seconds, the actual time is more than 40 second. The reason for this may be that the high index material after the completion of the interface cannot suddenly be stopped. The emission current can be immediately put to zero but the evaporation goes to zero after some time. The oscillating behavior of the index profile is due to the frequency variation of the Quartz crystals. Similar behavior is obtained for index profile vs. physical thickness of the interface which is in this particular case is ≈ 19 nm.

The results of our measurements on reflectance, absorption and damage threshold for the abrupt as well as gradual interfaces are listed in table 1. Comparing reflectivities of the four inhomogeneous AR-designs with abrupt systems, a clear trend reduction is visible. The design with Zirconia / Silica gives minimum reflectivity. A similar trend of detraction has been observed in absorption of inhomogeneous designs with the exception of Cerium oxide/Magnesium fluoride where an increase in absorption is apparent. Such absorption reduction in coatings with homogeneous or gradual interfaces can be explained on the basis of improvements in stoichiometric properties of the layer.

Table 1. The results show the inhomogeneous antireflection coatings.

Material	Reflection		Absorption		Damage Threshold {J/cm ² }	
	Abrupt	Gradual	Abrupt	Gradual	Abrupt	Gradual
HfO ₂ / SiO ₂	.15	.10	1072	219	42 ± 17	44 ± 9
ZrO ₂ / SiO ₂	.12	.08	1083	470	45 ± 10	51 ± 14
Ta ₂ O ₅ / SiO ₂	.39	.20	935	881	42 ± 6	49 ± 12
CeO ₂ / MgF ₂	.12	.11	940	984	37 ± 6	31 ± 6

These improvements have been noticed by Ristau [14] during the study of Auger electron spectroscopic (AES) measurements of $\lambda/2$ Titania / Silica systems in abrupt as well as in inhomogeneous coatings. Abrupt systems show a leakage of oxygen while coatings with gradual interfaces do not show any decrease of oxygen concentration at the interface.

The increase in damage threshold of three AR-2 oxide-oxide coatings could be due to an improvement in adherence at film-film interface. During experiments with abrupt AR-2 systems, morphological studies revealed in most of the AR-2 coatings that the upper layer is delaminated or peeled off. This observation suggests a poor adhesion at the film-film interface. Now, in the case of inhomogeneous coatings, adhesive properties are improved because of simultaneous evaporation of both materials. This means, the whole system behaves like a single film in which index of refraction varies continuously from high index to the low index material.

In contrary to oxides, the inhomogeneous system with CeO₂ / MgF₂ exhibit an increase in absorption. The reason for such a behavior could be a chemical reaction between oxide and fluoride in the mixed region which in turn increases the absorption and reduces the damage threshold. Such type of absorption enhancement in oxide-fluoride inhomogeneous systems have also been reported earlier [14]. These studies $\lambda/2$ systems of Titania and Tantalum in combination with Magnesium fluoride. Histograms of absorption and damage threshold have been presented separately in figure 2a & 4b for abrupt and gradual interfaces. It is apparent from the figures that systems which show a reduction in absorption in inhomogeneous layers also exhibit an increase in damage threshold. On the other hand, systems which show an increase in absorption, damage threshold is found to be decreased as compared to abrupt systems. Best result is obtained by Zirconia / Silica

(gradual interface) where damage threshold is improved from 42 (abrupt) to 51 (gradual) J/cm².

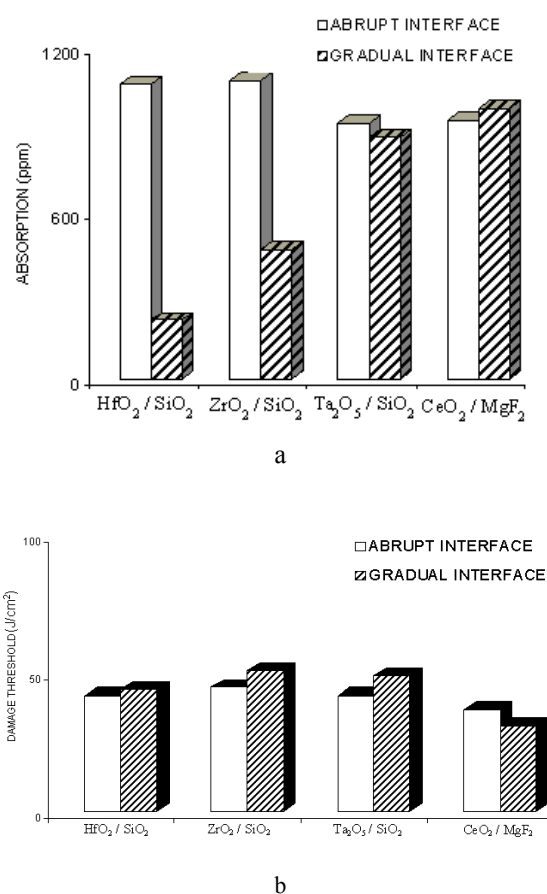


Fig. 4 a. Histogram of absorption for abrupt and gradual interface AR-systems b. Histogram of damage threshold for abrupt and gradual interface AR-systems

The three inhomogeneous designs consisting of both oxides exhibit a similar damage mechanism, i.e., removal of the upper layer and a uniformly melted portion in the middle due to absorption. But the delamination effect is not as strong as in case of abrupt systems which may be an indication of improved adhesion. AR-2 coating with Cerium oxide/Magnesium fluoride, both in abrupt and inhomogeneous case displays a similar type of damage, i.e., delamination and a uniformly melted portion in the high index layer.

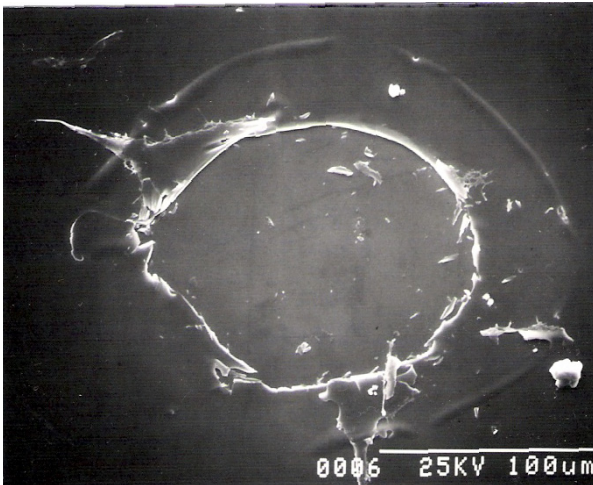


Fig. 5. Scanning electron micrograph of an inhomogeneous antireflection coating of Tantalum/Silica on Suprasil at 76 J/cm^2

The three inhomogeneous designs consisting of both oxides exhibit a similar damage mechanism i.e. removal of the upper layer and a uniformly melted portion in the middle due to absorption. But the delamination effect is not as strong as in case of abrupt systems. As an example, we show a damage site of Tantalum/Silica (codeposited) in scanning electron micrograph (Fig. 5).

4. Conclusions

In all the AR-systems discussed earlier [14], we have seen that high damage threshold coatings lie in the range of $40 - 46 \text{ J/cm}^2$. In order to improve further the damage thresholds of some best designs, AR-2 coatings were prepared with codeposited interfaces. In multilayer systems, most contribution of absorption loss comes from the interface. To reduce these losses it is noteworthy to produce coatings with gradual or codeposited interfaces. In oxide-oxide systems, a significant reduction in absorption and increase in damage threshold was observed. Best result was obtained by Zirconia/Silica coatings where damage threshold was improved from 42 to 51 J/cm^2 . On the other hand $\text{CeO}_2/\text{MgF}_2$ show an increase in absorption and decrease in damage threshold. This is in agreement with earlier results [14]. Damage morphology of these systems is the same as in case abrupt delamination effects due to the improvements in adhesion at film-film interface.

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