Very high damage threshold Al_2O_3/SiO_2 dielectric coatings for excimer lasers

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ABSTRACT

In the EUREKA EU205 project the target products are industrial excimer lasers in the average power range of one kilowatt or more. The high power optical components and dielectric coatings have to be developed in close adaption to cavity design (optics), beam relay optics, mask imaging optics, and masks. Therefore, we used ultra low loss conventional e-beam evaporation for Al_2O_3/SiO_2 dielectric multilayers. Based on a fundamental coating technique, both multilayer mean background absorption and absorption at localized spikes have been reduced drastically. The resulting KrF laser damage threshold of HR coatings is 16 J/cm^2 (1-on-1, 30ns, EMG-202-MSC). Measurements have been performed with an automated damage testing facility, being part of the EUREKA program. Multilayers have been characterized by Atomic Force Microscopy, Photothermal Microscopy, absorption measurements, and Spectroscopy of Sputtered Neutrals.

1. INTRODUCTION

Numerical design and manufacture of UV dielectric coatings are strongly influenced by the differing material properties in this wavelength region compared to the VIS and IR. Namely the short wavelength absorption edge is connected with higher binding energies that results in lower electronic polarizabilities. Consequently, there is a lack of extreme high index materials in the UV. Furthermore the number of UV transparent materials is limited. There are only a few high-low-refracting material combinations applicable.

Laser-induced damage threshold (LIDT) of the coatings is a limiting factor for the development of cost-effective excimer laser systems. A coating optimized UV beam delivery system reduces operating costs and increases system productivity. Coating parameter that most strongly influences LIDT is the choice of materials used to produce a component. UV-transparent materials of choice are several metal oxides/SiO_2 combinations (Al_2O_3, Y_2O_3, Sc_2O_3, and HfO_2), large bandgap fluoride combinations (MgF_2, Na_3AlF_6, LaF_3, NdF_3, GdF_3, AlF_3), and metal oxide/fluoride combinations.

Highest reported[1] LIDT values of KrF (248nm) reflectors (HR's) consisting of oxide layers are about 10J/cm^2. With the all fluoride LaF_3/Na_3AlF_6 248nm HR system non-conditioned 15ns LIDT of 13J/cm^2 was achieved[2]. The conditioned LIDT value of these systems is even 27J/cm^2. With the same fluoride material combination a 10ns LIDT of 20J/cm^2 has been achieved at 355nm HR's[3].

Optical stability investigations on oxide and fluoride thin-films are concentrated on the excimer wavelength (193nm, 248nm, 308nm, 351nm) and on the 3rd and 4th harmonic of the Nd-YAG laser (266nm, 355nm). They are classified about absorption, scattering, and laser damage threshold[4]. It has been shown that the microstructural sizes in fluoride thin-films are in the order of the UV wavelength scale. Therefore, structure related properties result in a variety of performance limiting effects. The observed optical inhomogeneities[5-8] below 100nm fluoride film thickness have been attributed to substrate-thin-film transition layers. It is of primary interest to note that the UV-quarterwave thicknesses fall directly into that undefined transition region.

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To overcome the well-known[9] problem of water uptake in thin films, alternative energy assisted deposition methods like sputtering, ion plating, ion beam sputtering, and ion assisted deposition have been used[10-12]. Up to now, the difficult problem of minimizing optical losses below 300nm wavelength remains. Classical and plasma enhanced CVD methods (PICVD, PECVD) are hopeful methods for producing stable UV films. However, the today's process temperatures are too high. State of the art deposition technology remains unsupported PVD on heated substrates under clean vacuum and substrate conditions. Interference design should be as simple as possible.

We concentrated on the development of Al_2O_3/SiO_2 coatings for KrF excimer lasers. This paper presents results about LIDT values and combined optical and nonoptical characterization of the coatings.

2. COATING PREPARATION

Coating has been performed in a BALZERS BAK 640 coating chamber equipped with a refrigerator cryopump and a LN_2 cooled Meissner trap. The coating and substrate cleaning equipment was under class 10.000 - 1.000 (particles/ft^3) clean room conditions. Film thicknesses and deposition rates were controlled by an optical monitor at 248nm and a quartz oscillator, respectively. Both Al_2O_3 and SiO_2 have been evaporated reactively from electron beam evaporators (6kV, 15kW total power) with rotary crucibles and Mo-liners. Substrate temperature was about 570K. No further energy input by ion- or plasma sources was made to avoid additional absorption losses. The codeposition of impurities must be carefully avoided since even traces of foreign material can cause serious optical losses and catastrophic laser damage.

Coating design was simply S | $(HL)^{20}$HLL | Air, with H: quarterwave layer Al_2O_3 (99.5% Patinal), L: quarterwave layer SiO_2 (99,89% Patinal), and S: substrate SQ1 (Glaswerke Jena) fused silica.

For comparison we have deposited 248nm HR coatings in a conventionally oil diffusion pumped vacuum chamber with LN_2 baffle, but otherwise identical conditions. In the following we call these samples *normal coatings* in contrast to the *so-called high purity coatings.*

3. CHARACTERIZATION

Characterization techniques used can be divided into optical and nonoptical techniques. The optical techniques are summarized in Table 1. Table 2 shows the nonoptical techniques.

UV-VIS spectrofotometer (190nm-900nm)	spectral characteristics, refractive index, thickness
damage testing facility	"1-on-1"-LIDT at 248nm, 30ns
laser calorimeter	integral absorption at 248nm
micrometer resolved photothermal displacement technique (PDT)*)	photothermal inhomogeneities

Table 1: Optical characterization of the EU205 coatings
*) in corporation with the Freie Universität Berlin

Scanning Electron Microscopy (SEM)**)	structural features
Atomic Force Microscopy (AFM)	surface topography
Spectroscopy of Sputtered Neutrals (SNMS)*)	chemical composition, impurities, H_2O content

Table 2: Nonoptical characterization of the EU205 coatings
*) in cooperation with the Jenoptik GmbH Jena
**) in cooperation with the IPHT Jena

4. RESULTS AND DISCUSSION

4.1. Damage thresholds

Single pulse damage thresholds of the deposited high reflection coatings have been measured in the UV damage testing facility of Laser-Laboratorium Göttingen, following ISO standard 11254 [13]. A detailed description of the used setup has been given elsewhere[14,15].

The main parameters used during the measurements are compiled in Table 3:

Laser:	EMG202 (Lambda Physik)
Wavelength:	248nm
Pulse duration:	30ns
Angle of incidence:	0 deg
Pulse energy variation:	variable attenuator (edge band dielectric filter)
Beam profile on target:	flat-topped
Effective beam size:	420µm x 250µm
Spacing of tested sites:	2mm

Table 3: Relevant parameters used for LIDT measurements

The flat topped beam profile is achieved by imaging a circular aperture with a spherical lens of 250mm focal length. The deviations on the plateau are about 20%, as monitored by a high resolution spatial beam profiling system[16].

High sensitivity on-line damage detection is performed by the help of a video microscopy system including digital image processing: The video micrographs of the site under test are acquired and stored before and after laser trigger; subsequent pixelwise comparison of the digitized images guarantees an unbiased decision whether or not the respective laser pulse has caused damage. The sensitivity of this technique is equivalent to off-line Nomarski microscopy[17], which is used for comparison.

Actual damage thresholds are determined within an automated measuring cycle, including sample positioning, laser triggering, on-line pulse energy measurement, damage detection, and threshold evaluation.

In Table 4 the obtained "1-on-1" damage thresholds are compiled for various high purity coatings deposited with slightly different process parameters. For comparison we have included also some normal coatings.

no.	sample	no. of layers	damage threshold [J/cm^2]
21	high purity	41	10.4
22	"	41	12.8
25	"	41	15.9
328/8	normal	49	2.48
365/5	"	41	2.8
SQ1	fused silica	-	3.51

Table 4: "1-on-1" damage thresholds of Al_2O_3/SiO_2 248nm HR coatings and fused silica substrate measured at 248nm (cf. text)

As seen from Table 4, damage thresholds of almost 16J/cm^2 are achievable in the high purity deposition regime, representing a dramatic increase in performance as compared to the coatings deposited under normal conditions. It is also interesting to note that the damage thresholds of the high purity coatings can be more than a factor of 4 higher than those of the used bare fused silica substrates.

4.2. Spectral performance

Fig. 1 shows the measured transmittance and transmittance + reflectance spectrum of a high purity Al_2O_3/SiO_2 248nm HR coating. Optical losses (scattering + absorption) are about 1% at 248nm and increase towards shorter wavelength due to the short-wavelength absorption edge of the metal oxides and increasing scatter losses. The spectral performance of the HR system does not give explanations about the very high LIDT. It shows only the exact spectral position of the system. Furthermore, the spectral position of short-wavelength absorption edge of the system suggests complete oxidation of the layers.

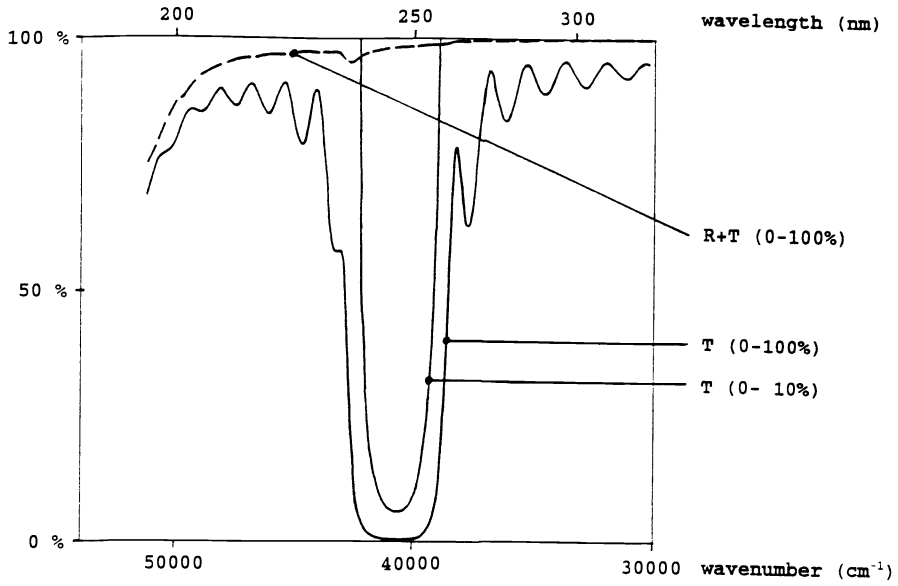

Fig. 1: Transmittance and transmittance + reflectance spectrum of a high purity Al_2O_3/SiO_2 HR coating S | $(HL)^{20}HLL$ | Air

4.3. Absorption

An UV-laser calorimeter currently under development at Laser-Laboratorium Göttingen was employed for a determination of the samples' absorptance at 248nm, using the "pulsed irradiation method" proposed in ISO/DIS 11551 [18]. The arrangement is briefly described in the following:

Samples are mounted inside a large aluminium chamber that provides decoupling from changes in the ambient temperature and will absorb stray light without considerable temperature rise. Radiation induced temperature increase of the sample is measured with a precision NTC-thermistor attached to the sample holder. Two precision 100µA current sources drive the NTC and a programmable reference resistor. The resulting voltage difference (sensitivity about 45mV/K at 0.1mW heat dissipation of the NTC) is amplified by an instrumentation amplifier with a gain up to 900. This signal is fed to a 14-bit A/D-converter within a Personal Computer.

The PC also controls an adjustable oscillator (max. 150Hz) which triggers the KrF laser. A variable attenuator is used to adjust the average laser power P_{av}. The sample is placed in the convergent beam behind a spherical quartz lens (f=750mm), with a slight inclination with respect to the incoming beam to guide the reflected beam out of the aluminium vessel onto a beam dump.

Fig. 2 shows two characteristic unfiltered temperature vs. time curves obtained for an irradiation time t_{irr}=30s. It is assumed that once the temperature distribution has become homogeneous, the sample cools exponentially. Thus the theoretical temperature rise ΔT (i.e. without thermal losses) is extrapolated from the cooling curve and, with the total laser energy $P_{av} \cdot t_{irr}$ and the heat capacitances of the sample and the mount, the absorptance A is computed.

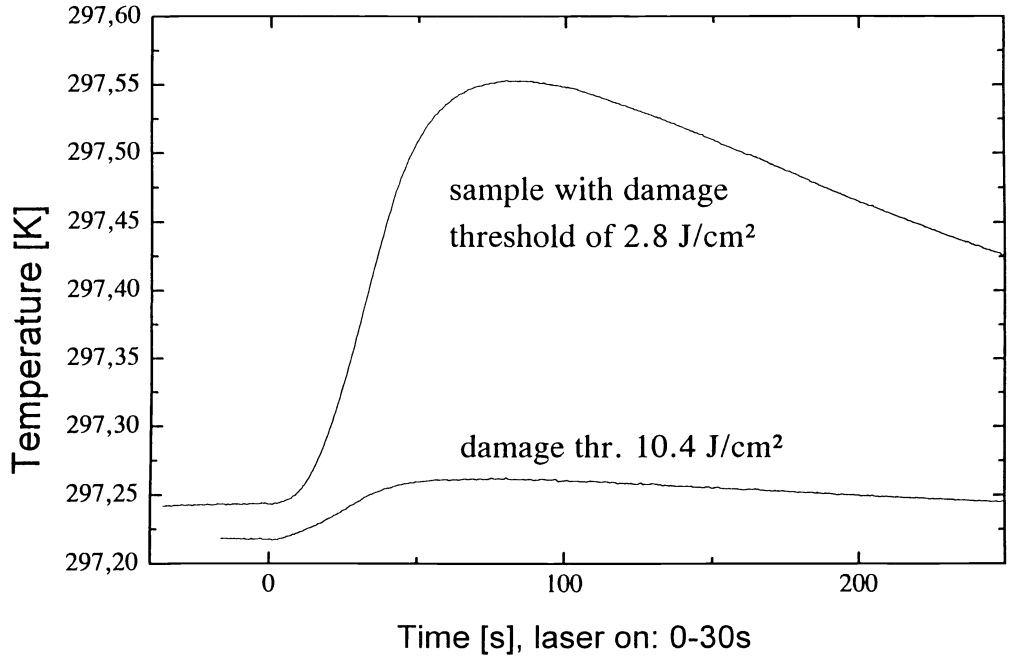

Fig. 2: Temperature vs. time curves for two laser irradiated HR coatings deposited under normal and high purity conditions, respectively; laser parameters: wavelength 248nm, pulse length 30ns, repetition rate 150Hz, pulse energy density 175mJ/cm2; respective damage thresholds are shown

Table 5. displays absorption data measured at pulse energy densities of 175mJ/cm^2; measurements at 25 and 120mJ/cm^2 yield similar results. Since the apparatus is still in its experimental stage and a precision calibration has not yet been performed, the absolute error may be in the order of 40%; the relative error, however, is much smaller.

no.	sample	damage threshold [J/cm^2]	absorption [%]
21	Al_2O_3/SiO_2	10.4	0.068
22	"	12.8	0.06
25	"	15.9	0.077
328/8	"	2.48	1.04
365/5	"	2.8	0.467
272/7	HfO_2/SiO_2	1.23	0.306

Table 5: Comparison of damage threshold and absorptance data of characteristic samples measured at 248nm

For the high purity HR coatings (#21, #22, #25) absorptance values below 0.1% are determined, which is well correlated to the respective high damage thresholds. Vice versa, the less damage resistant samples deposited under normal conditions exhibit much higher absorption data.

Comparing these data with the spectrometrically determined optical losses for the high purity coatings ((1-R-T), cf. Sect. 4.2.), a relatively high contribution of scattered UV light (about 10 x higher than absorbed) can be estimated.

Since a theoretical treatment of the thermally induced damage mechanism predicts an A^{-1}-dependence for the threshold fluence[19], a corresponding plot is shown in Fig. 3. Although there are only a few data points and the scattering is not neglectable, a linear relationship according to the theory is indicated. The data are also in qualitative agreement with results published by Itoh et al.[20], who used an interferometric method for the absorption measurement of HR oxide coatings.

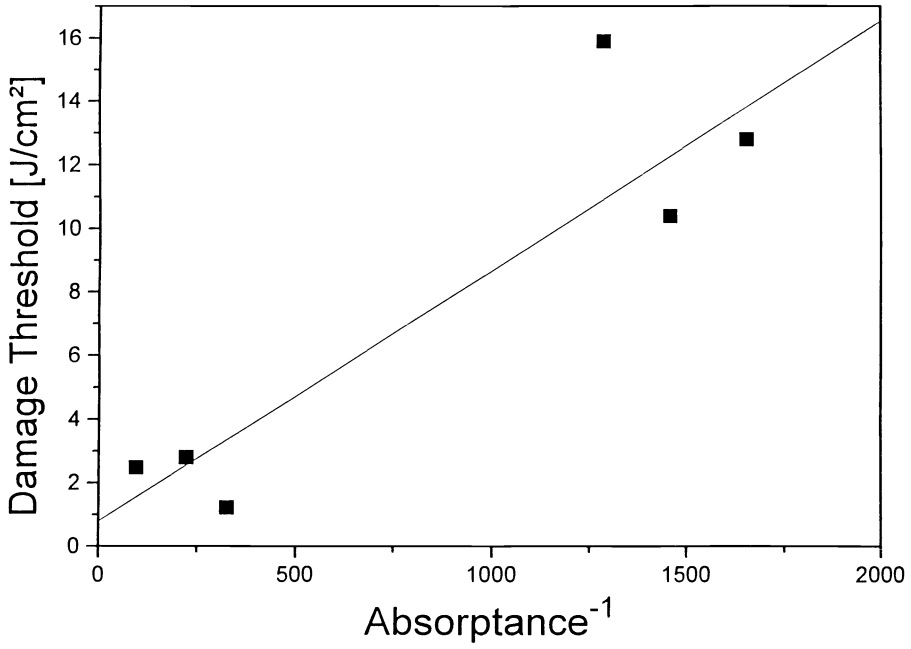

Fig. 3: Damage thresholds vs. inverse of absorptance, laser parameters as in Fig. 2

4.4. Photothermal displacement technique

CW photothermal displacement technique (PDT) has been shown to be a tool for the characterization of optical coatings with high spatial resolution combined with an ultrahigh sensitivity[20,21,22]. By micrometer resolved PDT-measurements we found that the non-damaged normal deposited thin film systems (#365/2) contain a great amount of photothermal inhomogeneities (defects) with lateral sizes ranging from several µm to several ten µm that are not visible by optical microscopy[22]. These areas of strongly enhanced displacement response should be originated from absorption centres rather than from thermal inhomogeneities in the film, from microdelamination and/or decreased thermal impedance at the film interfaces. High purity coatings (#25) show one order of magnitude lower concentration of photothermal defects and a significant lower photothermal background signal. These results (details see [22]) are in correspondence with the measured laser damage thresholds and absorptance of the HR coatings (cf. Tables 4 and 5).

4.5. SNMS, AFM, TEM

SNMS (5keV Ar^+ ions) has been used to determine the depth dependent impurity content in both normal and high purity 248nm HR coatings. To get constant O- signals. Among the impurity neutrals (O, OH, C, N) registered during sputter process, there were no differences in the impurity content between the normal and high purity film types for O, OH, N. However, the C- content does differ significantly.

In Table 6 the relative C-contaminations are compiled for various coatings deposited under normal and high purity conditions. For comparison we have included also a 248nm HR coating which has been deposited from a common coating supplier under non-cryo vacuum conditions. We call these sample *reference coating.*

no.	sample	no. of layers	relative integral SNMS C intensity [cps]	damage threshold [J/cm^2]
2	high purity	41	8	9.17
1	"	41	8	9.34
G	reference	43	8	4.34
328/2	normal	41	24	3.6
SQ1	fused silica (substrate)	-	8	3.51

Table 6: Relative integral SNMS C intensity of Al_2O_3/SiO_2 248nm HR coatings and fused silica substrate and "1-on-1" damage thresholds measured at 248nm

As seen from Table 6 the relative C signals of high purity and medium purity coatings are in the order of the background signal from the bare substrate and are not correlated to the damage thresholds. The normal coating shows a significant higher C-signal.

With the help of TEM of C/Pt replicas of film fracture edges the surface and volume structure of the HR coating was resolved. Between the normal and high purity coatings no difference in film microstructure is seen.

The top layer structure has been investigated by AFM. Within the uncertainties of the measurement, following the TEM result, no difference in grain size and surface roughness exists between normal and high purity coatings.

5. CONCLUSIONS

It has been shown that damage thresholds of almost 16J/cm^2 of Al_2O_3/SiO_2 248nm HR coatings in a high purity deposition regime are reproducibly achievable. Damage thresholds are well correlated to absorptance values at 248nm and photothermal displacement signals. A correlation to the C-contamination was only found at a sample of very low damage threshold.

Oxide coating raising of LIDT requires substance-specific knowledge about layer-structural factors of very large complexity[23]. However, it came out, that LIDT is due to nano- and microdefects (inclusions or point defects[24]) causing a homogeneous background absorption and macrodefects (dust particles trapped in the film or/and at the interfaces) causing localized absorption spikes[25].

Influence of macrodefects on LIDT has been investigated[25,26]. LIDT interpretation in terms of point defects[24] is strongly connected with thin film morphology and the low thermal conductivity of the coatings[27-30]. In our oxide coatings point defects are also possible first by empty oxygen vacancies or by carbon atoms or hydrocarbon molecules from the residual gas and vacuum chamber.

Because of the complexity of LIDT and the measurements undertaken, we are not able to clear all reasons for low and high LIDT up to now. However, under low macrodefect conditions the influence of hydrocarbons[31] and oxygen on LIDT is evident. Further investigations are necessary and coatings from other laboratories shell be tested in our LIDT facility and vice versa.

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DISCUSSION

Q- There are a couple of ways to do SNMS. Did you use an external ion source to etch the surface of the coating or were you able to see enough conductivity induced in the film to self bias it with respect to the plasma?

A- We used an ion source. It was not possible to use an argon ion source but a krypton source worked. It is difficult, you have so many artifacts that you have to work very long to get results that are real. We used a low voltage plasma and special care for fluoride and oxide films.

Q- What sort of spatial resolution do you have with SNMS, particularly in regions where you were seeing high absorption?

A- The problem is to get quantitative results with SNMS. You must use another substrate, not the original optical substrate. Then you are able to make a calibration of the carbon in the original substrate. We have done it with the fluorides but not yet with the oxides. You can resolve about 1/4 to 1/2 of the depth of a quarter wave layer.