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Investigation of laser-induced damage at 248 nm in oxide thin films with a pulsed photoacoustic mirage technique

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Abstract: Laser damage thresholds at 248nm of TiO_2, ZrO_2 and HfO_2 thin films of λ optical thickness on SQ1 quartz glass substrates are determined by the photoacoustic mirage technique. Damage thresholds correlate with the band gap energy of these materials, as determined by optical spectroscopy. It is demonstrated that the damage resistance can be raised by an additional $\lambda/2$ SiO_2 overlayer. Damage thresholds are identical for polycrystalline and amorphous film structure and not influenced by a change of substrate material from quartz to BK7 glass.

1. INTRODUCTION

Measuring laser damage thresholds of dielectric thin film systems is of interest from a general point of view and of great importance for practical applications and the durability of optical components. Especially since the advent of pulsed high-power laser systems demanded excellent performance of optical components, the laser damage threshold became an important figure of merit for optical coatings made of multilayer stacks of dielectric thin films. Among various materials used for optical applications the refractory oxides play a prominent role due to their wide range of applicability[1].

The purpose of the present paper is twofold. *First*, the method of pulsed photoacoustic mirage deflection [2,3] is introduced as a very sensitive tool for measuring damage thresholds of dielectric thin films. Results obtained by this method are compared to damage thresholds determined by reflectance and photothermal displacement techniques. *Second*, damage results for irradiation with 248nm excimer laser light are presented for selected oxide thin films of λ optical thickness, namely TiO_2, ZrO_2 and HfO_2.

2. EXPERIMENTAL TECHNIQUES

Measurements were performed on a set of 24 oxide samples with λ optical thickness (λ=248nm) deposited by electron beam evaporation. Process conditions were adjusted either to obtain a polycrystalline or an amorphous film structure. Furthermore, each material was deposited on SQ1 quartz and BK7 glass substrates for comparison, and the series was duplicated in order to study the influence of an optically inactive $\lambda/2$ protective SiO_2 overcoating.

Three different techniques were applied to measure the laser damage threshold: the pulsed photoacoustic mirage deflection [2,3], the surface reflectivity after laser irradiation and the measurement of light absorption by cw photothermal displacement microscopy [4]. While the first method was applied in-situ during the damaging laser irradiation, the latter measurements were performed afterwards.

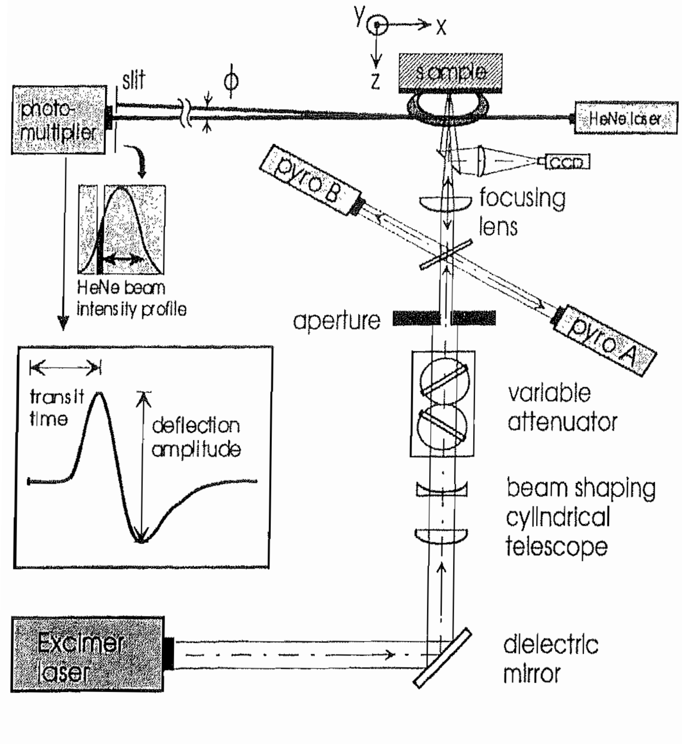

Fig. 1: Experimental scheme for pulsed photoacoustic mirage detection of laser induced damage.

A schematic overview of the mirage apparatus is given in Fig. 1. The damaging laser pulse from the excimer laser passes a variable attenuator to adjust the laser intensity and is focused on the sample surface into a spot of 450µm. Measurement of the incident laser pulse energy with a pyroelectric detector (pyro A) and of the beam profile by a CCD camera system allow an accurate determination of the incident laser fluence. All measurements were done in a 1-on-1 irradiation mode.

The transient change of the index of refraction in the ambient air, induced by the plasma-induced supersonic pressure wave leads to a mirage deflection of a probe laser beam aligned parallel to the sample surface. Data analysis of the mirage signal was restricted to deriving only deflection amplitude (peak to peak) and transit time, i.e., the time elapsed between the laser pulse and the arrival of the acoustic pulse at the location of the probe laser beam (cf. Fig. 1).

Reflectance and surface displacement were measured in another apparatus by scanning a probe laser beam (HeNe-laser, 632nm) over irradiated spots. For displacement measurements, the local thermoelastic deformation resulting from heating by a modulated pump beam (Ar$^+$-ion laser) was probed by the change in reflection angle of the probe beam, measured by a quadrant photodetector (Fig. 2, left). The average value of the resulting displacement amplitude profile (Fig. 2, right) was taken as a measure of the film absorption. For reflectivity measurements the pump beam was switched off and the total reflected probe beam intensity integrated over the two-dimensional profile represents the reflectance signal.

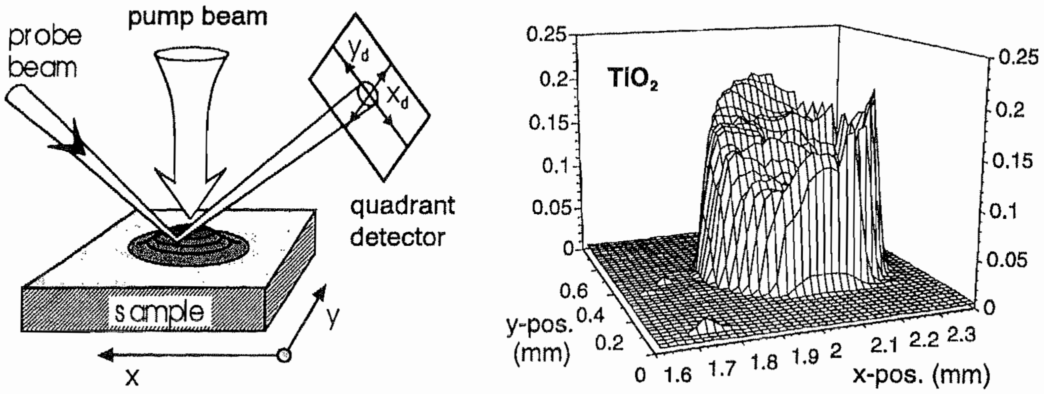

Fig. 2: Principle of photothermal displacement detection (left). Two-dimensional displacement scan of laser damaged area in a TiO_2 thin film (right).

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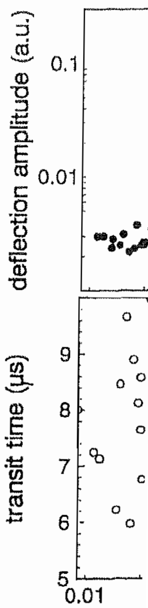

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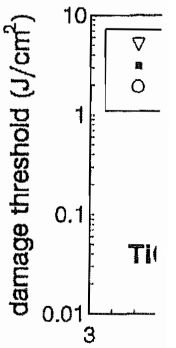

Fig. 4: Correlati
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3. DAMAGE THRESHOLDS

All signals were recorded as a function of the stepwise increased excimer laser fluence. Typical results for TiO_2 obtained by the techniques described above, are displayed in Fig. 3. A distinct threshold behaviour was found in all curves, however, the lowest threshold (most sensitive measurement) is recorded by the mirage effect. Therefore, this technique is used as the standard method for threshold determination.

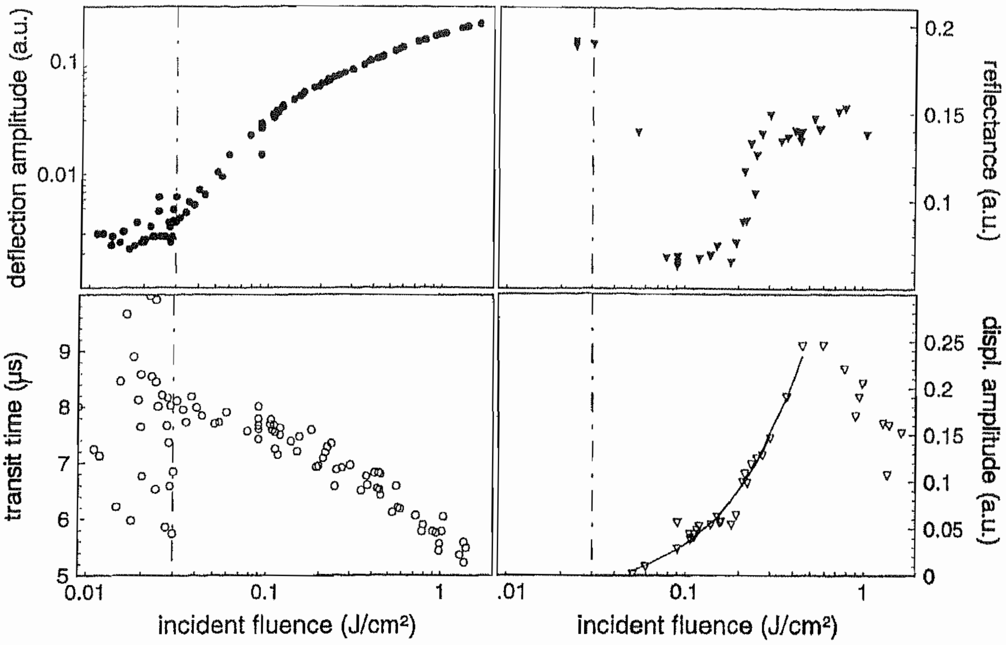

Fig. 3: Comparison of damage results obtained by three different techniques. As a function of incident fluence, the left side displays mirage data (c.f. Fig.1), the right side shows the reflectance at 632nm (upper) and displacement (lower, c.f. Fig. 2).

In Fig. 4 the thresholds of all oxides determined by the mirage method are releated to the band gap of the respective thin film material. It is found that the apparent threshold is strongly correlated with the gap energy, and data suggest a power law for the relation between both quantities. This dependence reflects the different levels of absorption of the 248nm laser light by the oxides. While the optical absorption edge is far above the excimer laser wavelength for TiO_2 it is just in the vicinity for ZrO_2 and below 248nm for HfO_2. A similar variation of the incubation was previously observed for these oxides. [5] However, the damage threshold is not solely determined by the intrinsic electronic properties of the thin film material but also by the thin film structure which in turn depends on the growth conditions [6]. Furthermore extrinsic thin film defects incorporated during the deposition process or resulting from environmental influences may have a major influence [7].

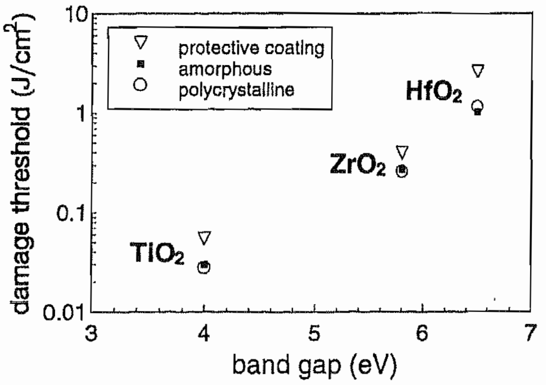

Fig. 4: Correlation between laser damage threshold for 5eV photons and band gap of oxide thin films.

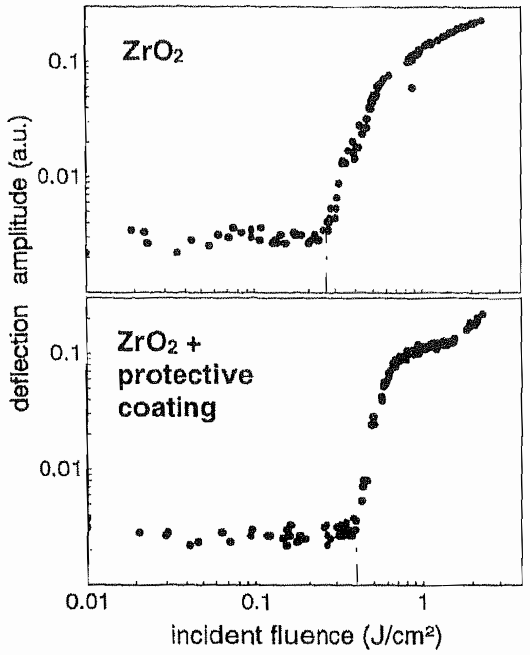

Fig. 5: Shift of damage threshold at 248nm for the ZrO_2 thin film by a SiO_2 protective coating.

In order to investigate the influence of various parameters on the damage threshold, measurements were carried out with the entire set of samples. It was found that thresholds are identical for samples with polycrystalline and amorphous structure. However, the deposition of an additional protective coating resulted in an improvement of damage resistance by a factor of two for all thin film systems. Results for ZrO_2 shown in Fig. 5 reveal that the overcoating does not only change the threshold but also the nature of the damage process. While the fluence dependent mirage deflection shows a moderate increase above threshold for the single film system, a much steeper slope was found for the sample with the additional protective coating. The lower trace also shows a step above $1 J/cm^2$ that might be interpreted as a second threshold appearing after removal of the first layer. Similar results were found for the other oxides.

In another set of experiments damage thresholds of oxide films deposited on SQ1 quartz were compared to those from films on BK7 glass. The thresholds were found to be the same regardless of the substrate material. Although, most of the excimer laser pulse energy is deposited in the glass, the resulting temperature rise is not high enough to influence the film damage. The reasons are that, first, the absorption coefficient of BK7 is much smaller than that of the oxides and, therefore, the heat is spread over a large volume. Second, due to the thin film thermal properties and a possible thermal contact resistance at the interface the heat does not penetrate deeply into the oxide film. Third, thin film defects can be detrimental to the film resistivity and dominate over substrate absorption effects.

4. ACKNOWLEDGEMENT

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5. REFERENCES

[1] Macleod H.A., Thin-film optical filters (2nd ed., Adam Hilger Ltd., Bristol, 1986) pp. 504-508.

[2] Petzoldt S., Elg A.P., Reichling M., Reif J., Matthias E., "Surface laser damage thresholds determined by photoacoustic deflection", *Appl. Phys. Lett.* **53**(21) (1988) 2005-2007

[3] Sell J.A. and Heffelfinger D.M., "Photoacoustic and photothermal beam deflection as a probe of laser ablation of materials", *J. Appl. Phys* **69**(3) (1991) 1330-1335

[4] Welsch E. and Reichling M., "Micrometer resolved photothermal displacement inspection of optical coatings", *J. Mod. Opt.* **40**(8) (1993) 1455-1475

[5] Matthias E. and Wu Z.L., "Non-Destructive Reading of Laser-Induced Single-Shot Incubation in Dielectric Coatings", *Appl. Phys.* **A56** (1993) 95-98

[6] Hacker E., Lauth. H., Weißbrodt P., Wolf. R, "Structural influences on the laser damage thresholds of oxide coatings", *Proceedings Europto series SPIE Vol. 1782* (1992) 447-458

[7] Bodemann A., Kaiser N., Reichling M., Welsch E., "Micrometer resolved inspection of defects and laser damage sites in UV high-reflecting coatings by photothermal displacement microscopy", paper presented at this conference

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