Available online at www.sciencedirect.com

ScienceDirect

Optics Communications 272 (2007) 221–226

OPTICS COMMUNICATIONS

www.elsevier.com/locate/optcom

Analysis of material modifications induced during laser damage in SiO_2 thin films

L. Gallais *, J. Capoulade, F. Wagner, J.Y. Natoli, M. Commandré

Institut Fresnel (UMR CNRS), Ecole Généraliste d'Ingénieurs de Marseille, Université de Provence, Université Paul Cezanne,
Domaine Universitaire de St Jérôme, 13397 Marseille Cedex 20, France

Received 21 August 2006; received in revised form 13 November 2006; accepted 15 November 2006

Abstract

The damage mechanisms in silica thin films exposed to high fluence 1064 nm nano-second laser pulses are investigated. The thin films under study are made with different techniques (evaporation and sputtering, with and without ion assistance) and the results are compared. The material morphological, optical and structural modifications are locally analyzed with optical microscopy and profilometry, photoluminescence and absorption microscopies. These observations are made for fluences near and above the laser damage threshold, and also in the case of multiple pulse irradiations. An increase in absorption in and around the damages is observed, as well as the generation of different defects that we spatially resolve with absorption and luminescence mappings.

Keywords: Laser induced damage; Optical coatings; Silica

---

1. Introduction

Laser induced damage in optical materials is a key issue for high power laser applications, and a significant number of studies have been published since the advent of lasers. It is widely acknowledged that the damage initiation in the nano-second regime on the surface of wide band-gap materials such as silica, is linked to the presence of nano to micrometer sized defects. These nanometric laser damage precursors may originate from the manufacturing processes (cleaning, polishing, coating) [1–3]. The study of the laser damage initiation mechanisms is difficult, mainly because the identification of nanometric laser damage initiators with low density on silica surfaces is still an issue. To overcome this problem, one solution is to intentionally introduce laser damage initiators and study their behaviour under laser irradiation. Progress has recently been made in the understanding of the laser damage process by studying artificial defects of known size and composition, embedded in silica thin films [4,5]. However the mechanisms could be very different on 'real' samples. Due to the low surface density of the damage precursors, investigations on real samples are usually based on the analysis of already damaged sites [6–9].

For this work we define the 'Damage Threshold' of the sample as the lowest fluence at which we observe mechanical damage of the sample by Normarsky microscopy. We describe here observations of a very early state of laser induced damage, that often is not detected. Using optical profilometry, absorption mappings and luminescence mappings, we also detected laser induced material modifications on 'undamaged' sites (according to the definition above). We will summarize these modifications by the word 'pre-damage'. The study has been carried out on bare fused silica substrates and on silica coatings deposited on these substrates.

---

[*] Corresponding author. Tel.: +33 4 91 28 80 72; fax: +33 4 92 28 80 67.
E-mail address: laurent.gallais@fresnel.fr (L. Gallais).
URL: www.fresnel.fr (L. Gallais).

0030-4018/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.optcom.2006.11.025

2. Experiment

2.1. Samples

The SiO_2 thin films (200 nm physical thickness) were prepared at the Fresnel Institute by different techniques: electron beam deposition (EBD), reactive low voltage ion plating (IP), dual ion beam sputtering (DIBS) and ion assisted electron beam deposition (IAD). The substrates are Herasil samples especially polished for high power applications, and originating from the same production and polishing batch. Prior to damage testing, all samples have been cleaned with an automatic aqueous cleaning procedure involving ultrasonic immersion and detergents followed by de-ionized water rinsing and drying. Details about the manufacturing of these samples can be found in reference [10].

The *E*-field distribution of the system silica substrate – SiO_2 air is not taken into account in the interpretation of the results since the refractive index of two materials are very closed.

2.2. Laser damage measurements

We irradiated the samples with a Nd:YAG laser (1064 nm, with a pulse duration of 6 ± 1 ns) tightly focused onto the surface (12 μm diameter at $1/e^2$). The test apparatus used for this purpose is described in detail in reference [11]. Laser damage is detected by Nomarski microscopy at 50× magnification. An image of the irradiated zone is acquired by a CCD camera before and after each shot. A simple image processing algorithm then detects changes due to the laser shot. We perform statistical measurements by irradiating 50 sites for each tested fluence and 20 different fluences are tested. Thus we have created on each sample a matrix of 1000 isolated sites irradiated with single shots at different fluences, ranging from fluences that do not result in any modification visible by conventional microscopy, to fluences inducing large damages at the surface of the material. Indeed, laser damage being stochastic by nature due to the initiation by defects of low surface density, we have to test a large number of sites to be able to observe the different steps of damage development. It is then possible to observe the evolution of the irradiated surface under increasing fluences with the different diagnostic tools described below.

2.3. Morphological measurements

The morphological changes have been observed with a surface profiler (Talysurf CCI 3000 Å), with a vertical resolution better than 0.1 nm and horizontal resolution of 0.5 μm in order to measure the dimensional characteristics of the damaged sites. Optical surface profilometry has been associated with Nomarski microscopy (Carl Zeiss Axiotech microscope).

2.4. Absorption and luminescence measurements

Absorption measurements have been performed using a photothermal deflection set-up with high lateral resolution. This 'photothermal microscope' enables the acquisition of absorption mappings at 1064 nm with micronic resolution [12]. Small isolated absorbing defects can be detected with this technique down to a limit of 50 nm diameter gold inclusions in silica [13].

Luminescence mappings have been acquired simultaneously with scattering and lower resolution absorption mappings all excited by a 244 nm pump beam [14]. The luminescence spectrum of isolated (micronic) defects can also be recorded with this apparatus: the minimum excitation spot diameter is 3 μm.

In this paper we report only relative variations of absorption and luminescence. For this reason the calibration procedure of these two apparatus will not be discussed, but more information about calibration and detection limit of the technique can be found in reference [15].

3. Results and discussion

3.1. Laser damage thresholds

The laser induced damage threshold (LIDT) for the coatings are dependent on the fabrication technique: they range from 12 J/cm^2 for EBD to 35 J/cm^2 for DIBS and IAD. For comparison, the front surface LIDT of the uncoated Herasil sample was 70 J/cm^2. The results are summarized in Table 1 and the details of the LIDT measurements can be found in reference [10].

3.2. Laser damage morphology slightly above LIDT

Despite the difference in the LIDT values, the same kind of laser damage morphology is observed on all coated samples at fluences only slightly higher than the LIDT: One or several deep pits in the layer (in most cases one, but in some cases two or three) (Fig. 1, detail A), and a shallow print reproducing the laser spot are observed (Fig. 1, detail B). For fluences 50% higher than the LIDT delamination of the layer occurs.

In case of the uncoated substrate, crater formation has been observed. The crater diameter is about 12 μm corresponding to the laser spot diameter.

Table 1
Comparison of the front face LIDT obtained on coated and uncoated Herasil samples (1064 nm, 6 ns, 1-on-1 mode)

Deposition technology	Substrate	EBD	IP	IAD	DIBS
LIDT at 1064 nm	70 J/cm^2	12 J/cm^2	20 J/cm^2	35 J/cm^2	35 J/cm^2

EBD: electron beam deposition, IP: reactive low voltage ion plating, IAD: ion assisted electron beam deposition, DIBS: dual ion beam sputtering. The coated layer was 200 nm thick SiO_2.