LASER DAMAGE AND ABLATION OF DIFFERENTLY PREPARED CaF_2(111) SURFACES *)

J. SILS **)†)

Institute of Solid State Physics, University of Latvia, 8 Kengaraga Str., 1063 Riga, Latvia

M. REICHLING, E. MATTHIAS

Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

H. JOHANSEN

Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany

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Ablation thresholds and damage behavior of cleaved and polished CaF_2(111) surfaces produced by single shot irradiation with 248 nm/14 ns laser pulses have been investigated using the photoacoustic mirage technique and scanning electron microscopy. The standard polishing yields an ablation threshold of typically 20 J/cm^2. When surfaces are polished chemo-mechanically the threshold is raised to 43 J/cm^2. Polishing by diamond turning leads to intermediate values around 30 J/cm^2. Cleaved surfaces possess no well-defined damage threshold. The damage topography of conventionally polished surfaces shows ablation of flakes across the laser heated area with cracks along the cleavage planes. In the case of chemo-mechanical polishing only a few cracks appear. Diamond turned surfaces show small optical absorption, but cracks and ablation of tiles. The origin of such different damage behavior is discussed.

I Introduction

Laser damage and ablation of transparent dielectrics has been an important area of study for many years [1–4]. Among the materials used in optical applications, CaF_2 plays a prominent role for handling excimer laser radiation, specifically for advanced UV-laser lithography where optical elements have to meet the highest standards with respect to both, optical quality and damage resistivity [5]. The latter has been investigated for CaF_2 in particular with 248 nm — a wavelength today used in laser lithography — and was found to depend sensitively on the surface quality [6]. In our previous studies we measured the damage and ablation behavior of conventionally polished CaF_2 (111) surfaces [7,8] and presented a first comparison with surfaces prepared by advanced polishing techniques such as diamond turning and chemo-mechanical polishing [9]. Scanning electron microscopy (SEM) was utilized for surveying irradiated spots and characterizing their damage

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**) Corresponding author; e-mail: sils@latnet.lv

†) Also at *Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany.*

topography [10]. In the present work we study ablation thresholds and damage features of samples treated with advanced polishing techniques. We demonstrate that with these techniques one is able to prepare surfaces that can withstand about five times higher laser fluences than those investigated in our earlier studies mentioned above. The ultimate goal is to explore possibilities for further increasing the laser damage resistivity and approach the value of the perfectly flat terrace on a cleaved crystal.

II Experimental

Three types of differently polished and one cleaved $CaF_2(111)$ surface have been investigated. The first sample (A) met usual optical standards and was polished by a conventional technique using diamond grit. The surface of sample (B) was processed by diamond turning, a technique where a sharp diamond tip is moved across the surface to remove a thin layer of material [11]. Sample (C) was chemo-mechanically polished [12] and cleaned by subsequent ion milling. The (D) surface was obtained by cleaving a single crystal under normal conditions. All surfaces were irradiated with 248 nm/14 ns laser pulses and their response was detected by the photoacoustic mirage technique, described in detail elsewhere [13,14]. In short, an acoustic wave travelling with the speed of sound is generated when the surface is heated above the ambient temperature. At higher fluences, once a plasma is formed, a supersonic shockwave develops for which speed and amplitude increases with increasing fluence.

The amplitude and arrival time of the mirage signal, the transmission of the sample, as well as plasma light emission, were monitored as a function of the incident

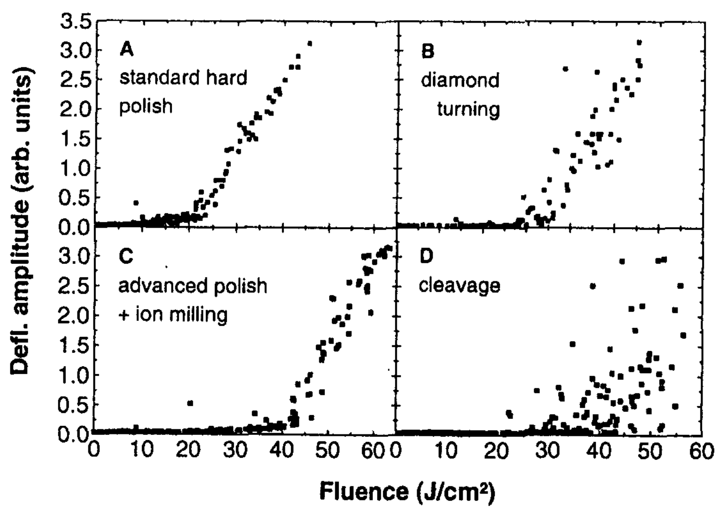

Fig. 1. Fluence dependence of the photoacoustic signal amplitude for differently prepared (111) surfaces of CaF_2 crystals. A) Conventional standard polish; B) prepared by diamond turning; C) chemo-mechanical polish; D) cleaved surface.

laser fluence. Here, we only discuss the mirage signal amplitudes displayed in Fig. 1 for the four surfaces which reveal sufficient information about pre-damage heating and the onset of ablation. All data points in Fig. 1 represent single shot results, i.e., each fluence value was obtained with one laser pulse directed at a previously not irradiated surface spot. Independent of surface treatment, no signal was detectable at fluences below $10\,J/cm^2$. When increasing the fluence, we observe a weak increase of the acoustic amplitude between $(10–20)\,J/cm^2$ for the standard polish (Fig. 1A) and $(20–40)\,J/cm^2$ for the advanced polish (Fig. 1C). The onset of ablation is marked by a sharp increase in amplitude generated by the shock wave. This ablation threshold is well defined for samples A and C, somewhat fuzzy for the diamond turned surface, and non-existent in the case of the cleaved surface. The reason for the latter is the varying density of steps within the laser spot size of typically $140\,\mu m$ when moving the laser beam across the surface. We have reason to believe that damage starts at steps.

Scanning electron microscopy (SEM) recorded in secondary electron mode was used as a complementary experimental technique which allows a visual inspection of the damage topography produced by high fluences. Based on this inspection we define the damage threshold as the lowest fluence value for that we observe cracks in the surface that is normally significantly lower than the ablation threshold. Sensitivity to surface charging also renders SEM capable of detecting minute material modifications even at fluences far below the damage threshold [10,15]. We made an effort to establish a correlation between the acoustic wave signals and the actual surface modifications generated by the laser shots.

III Results and discussion

Comparison of the acoustic data for all samples A through D in Fig. 1 shows a dramatic dependence of the threshold behavior on surface preparation. In Fig. 2 the damage topographies are shown for each of these samples after irradiating them with specific fluences stated in the figure caption. Albeit sold as an optical grade crystal, the surface polished by standard techniques showed the lowest ablation threshold of $20\,J/cm^2$ (see Fig. 1A). Below this value, surface heating occurs which we attribute to absorption of light by surface defects. The damage pattern above the threshold in Fig. 2A consists inside the spot of long cracks along (111) cleavage planes and massive removal of large tiles. In contrast, we notice at the periphery a high density of short cracks and no ablation. The reason is different thermoelastic forces acting inside and at the edge of the laser spot. Since we worked with a top hat laser profile, heating is fairly homogeneous across the spot. Hence, inside there exists a strong *vertical* temperature gradient but the lateral one is negligible. By exceeding the thermoelastic stress limit the vertical gradient causes a rupture at a certain depth parallel to the surface, leading to ablation of tiles. At the periphery of the spot, on the other hand, the strong lateral temperature gradient generates sheer stress along the surface which is responsible for the high density of short ranged microcracks, discernible at the periphery [7,8].

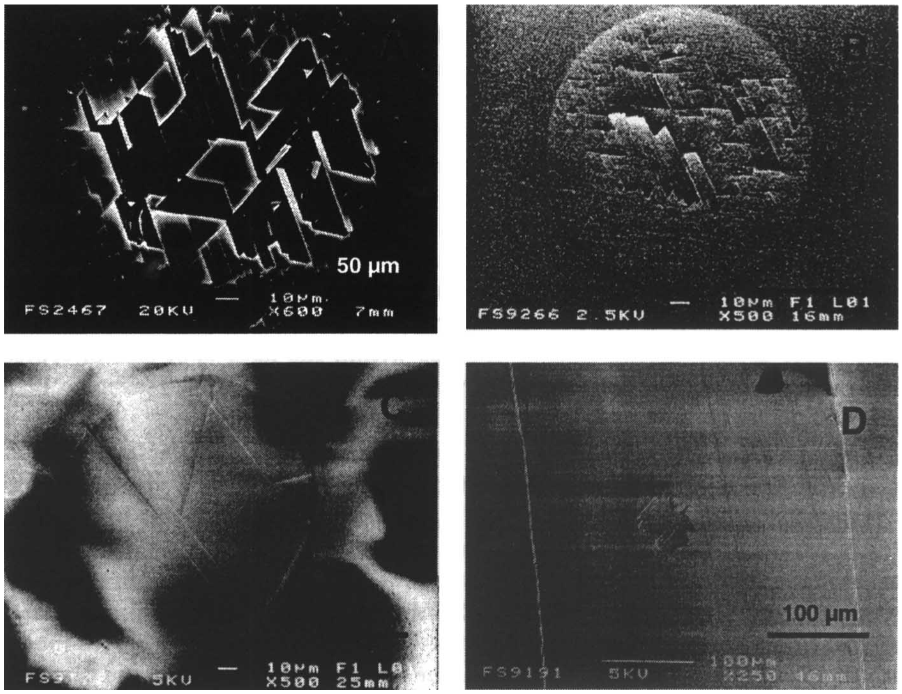

Fig. 2. SEM images of damage topographies of $CaF_2(111)$ surfaces irradiated with the following fluences: A) 27.6 J/cm^2 at the conventional standard polish; B) 30 J/cm^2 at the diamond turned surface; C) 57.7 J/cm^2 at the chemo-mechanically polished surface; D) 46 J/cm^2 at the terrace area of a cleaved surface.

A significant increase of the damage threshold can be achieved by diamond turning. We observe a well defined damage threshold around 28 J/cm^2 (Fig. 1B), about 40 % higher than the one for the conventional polish. For this polish we notice no apparent surface heating below threshold. The scatter of the data above the threshold indicates that damage seems to start rather localized and differs significantly from spot to spot. SEM inspection in Fig. 2B exhibits large size damage inside the spot but a surprisingly sharp boundary without any lateral cracks. The central damage pattern is reminiscent of the one in Fig. 2A, which means that damage still originates from the vertical temperature gradient. Missing cracks at the periphery, on the other hand, indicate that the high pressure applied during the turning procedure has destroyed the crystalline structure of the surface region. It seems that it produced an amorphous layer with little optical absorption in which no oriented cracks can develop under thermoelastic stress.

The chemo-mechanical polishing technique yielded the highest ablation threshold at a fluence value around 43 J/cm^2 (Fig. 1C). A number of SEM investigations with this sample revealed that mostly defects not removed by the polishing process initiate the damage. Interestingly, the overall damage behavior is different compared to the two previous cases. It is characterized by only a few straight fracture lines across the laser spot (Fig. 2C). In electron microscopy micrographs no material removal in form of tiles could be observed even at the highest applied fluences reaching up to $60\,J/cm^2$. The fracture lines observed by SEM seem to originate near the edge of the laser spot. In contrast to conventionally polished surfaces where damage starts at the place of highest vertical temperature gradient, here the highest lateral temperature gradient seems to trigger the cracks. Although there is a slight increase of the acoustic signal below the threshold (Fig. 1C), absorption inside the spot seems to be significantly reduced, indicating a low concentration of structural surface defects. As a consequence, vertical stress is too weak to overcome the thermoelastic stress limit.

Contrary to polished surfaces, the mirage signal of the cleaved surface does not reveal a well defined damage threshold (see Fig. 1D). Instead, over a wide fluence range we find a random distribution of acoustic amplitudes ranging from values close to the noise level to very high signals. Some spots remain undamaged even above $40\,J/cm^2$. This large scatter of the acoustic data is caused by the step density inside the laser spot. Cleavage steps are easily damaged while terraces are rather radiation resistant due to their low concentration of structural defects and absorption centers [16]. A rare example of a damage spot on a terrace is displayed in Fig. 2D. As can be seen from the length scale, in this case the laser spot was fortuitously placed on a terrace between two steps. The fluence was $46\,J/cm^2$, which obviously lies above the damage threshold of terraces. The source of this type of damage is not clear but most likely structural defects like dislocations or microsteps not resolved in this micrograph that give rise to the initial light absorption or weaken the surface.

IV Summary

We have compared single-shot damage produced by 248 nm/14 ns laser pulses on differently polished and cleaved $CaF_2(111)$ surfaces. Large differences were found in the laser resistivity of conventionally polished, diamond turned, and chemo-mechanically polished surfaces, and those prepared by cleavage. Conventionally standard polished surfaces exhibited the lowest thresholds, chemo-mechanically polished ones the highest. The resistivity of diamond turned surfaces came out in between. The fluence threshold for damage on a terrace was derived from an SEM image and found to be comparable to the one for a chemo-mechanically polished surface, indicating that this type of polish is superior. SEM also revealed different topographic damage features for the different polishing techniques. For the chemo-mechanically polished surface no ablation in form of tiles could be detected for fluences up to about $60\,J/cm^2$, damage only resulted in cracks across the laser spot. The other two polishing techniques produced surfaces that could be damaged by fluences between 20 and $30\,J/cm^2$ with ablation starting in the center of the spot and occurring in the form of regularly formed tiles.

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