We investigate phase imaging as a measurement method for laser damage detection and analysis of laser-induced modification of optical materials. Experiments have been conducted with a wavefront sensor based on lateral shearing interferometry associated with a high-magnification optical microscope. The system has been used for the in-line observation of optical thin films and bulk samples, laser irradiated in two different conditions: 500 fs pulses at 343 and 1030 nm, and millisecond to second irradiation with a CO$_2$ laser at 10.6 μm. We investigate the measurement of the laser-induced damage threshold of optical material by detection and phase changes and show that the technique realizes high sensitivity with different optical path measurements lower than 1 nm. Additionally, the quantitative information on the refractive index or surface modification of the samples under test that is provided by the system has been compared to classical metrology instruments used for laser damage or laser ablation characterization (an atomic force microscope, a differential interference contrast microscope, and an optical surface profiler). An accurate in-line measurement of the morphology of laser-ablated sites, from few nanometers to hundred microns in depth, is shown.

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1. INTRODUCTION

Laser-induced damage is defined by the ISO (International Organization for Standardization) standard as "any permanent laser-induced change in the characteristics of the surface of the specimen which can be observed by an inspection technique and at a sensitivity related to the intended operation of the product concerned" [1]. The damage threshold definition is then subjective and related to the detection method. As a consequence, many detection methods have been reported in the literature, with each one trying to give the most accurate threshold of the laser-induced damage occurrence. There are mainly two categories of detection methods: some are based on the detection of the material modifications, while others are based on the damage event detection.

As damage detection techniques in laser-induced damage threshold (LIDT) experiments, the most common, simple, and least expensive one at present time is the observation of the scattering coefficient of the specimen surface [2,3]. This scattering originates either from a probe beam or from the testing laser itself [3,4]. Simple to implement, this technique has been widely used. However, given the fact that the measured signal is exposed to a lot of noisy sources from the environment, it has to be complemented with other techniques such as off-line microscopy for reliable measurements [5,6]. Monitoring of transmission changes has also been reported [7]. The most common technique, which is recommended by the ISO standard [1], is optical microscopy, particularly in the differential interference contrast (DIC) mode. The interest in microscopy is that it combines damage morphology information to high sensitivity. This technique is mainly applied off-line but can be used in a laser damage experiment for in-line damage detection [8].

The damage detection techniques based on monitoring the damage event rely on the detection of the generated shock wave in air or in the material or on the detection of the plasma emitted in the damaged area. For example, the measurement of the photoacoustic effect can be done with a piezo sensor, an adapted microphone, or the deflection of a probe laser beam [9–12]. The monitoring of the transmission of the laser pulse itself was also proposed as a LIDT detection technique [13,14].

While looking for reliable and more sensitive techniques, the observation of the local damage gives better results and more information when techniques are compared on the same
experimental setup [15]. Microscopy in the DIC mode allows the detection of refractive index modifications in the material, birefringence effects related to stress, thickness differences related to delamination of a film, surface burning due to plasma, etc. However, only qualitative analysis can be done with this technique and if quantitative measurements are needed, they should be obtained with other off-line techniques (atomic force microscopy, scanning electron microscopy, optical surface profilometry, etc.). An alternative to obtain in-line quantitative data is to use a versatile quantitative phase microscopy technique that can be inserted into the laser-damage experimental setup.

In this paper, we use quadri-wave lateral shearing interferometry (QWLSI) as a wavefront sensor in order to provide quantitative phase microscopy as previously developed by Bon and coworkers [16]. We apply the technique to off-line as well as in-line measurements of laser-induced damage. Suitability, interest, and limitation of this new technique are compared to others for the field of laser materials interactions in general and laser damage in particular. After a description of the technique and the different experimental setups we have used, we will present LIDT measurements made with QWLSI. Thereafter, we will apply it to obtain quantitative information on laser-irradiated sites, such as the morphology of the ablated sites. To finish, we will apply the technique for the investigation of laser damage process in the case of an optical thin film irradiated at 500 fs, 343 nm. Comparisons with other measurements made with DIC microscopy, optical surface profilometry, or atomic force microscopy will be discussed all along the study.

2. MATERIALS AND METHODS

A. Quantitative Phase Imaging

With conventionally used DIC microscopy, also referred to as Nomarski microscopy, images present an output intensity that is a mix of amplitude and phase gradient contrast with a nonlinear response to optical path length gradient in the sample. Standard DIC systems are thus mostly qualitative in nature. That is why we propose to use a quantitative phase microscopy technique that has initially been developed to observe transparent living cells for biological applications [16]. There is much interest in this technique for multiple laser-damage applications: it is achromatic, self-referenced, and used as a standard camera on a microscope since any illumination (coherent or not, including white light) is possible. The wavefront sensor is placed in the image plane of the microscope to record the change in the wavefront shape that comes from the sample optical thickness and refractive index distribution [17]. This wavefront sensor is based on QWLSI [18,19]. QWLSI uses diffractive optics that makes four replicas of the incoming light beam to form an interference pattern onto a camera sensor (Fig. 1). A modified Hartmann mask (MHM) is used instead of the theoretically perfect but not available two-dimensional sinusoidal phase grating [20]. The two-dimensional interferogram contains information relative to the optical path difference gradients of the beam along both x and y spatial directions. Then it is possible to integrate these gradients in order to recover the actual OPD map and intensity images of the sample through Fourier analysis of the interferogram [21]. Integration of the diffractive optics just a few millimeters in front of the camera sensor is the key for the simplicity of implementation of the system, but the price is the reduction of the image resolution. For instance, the intensity and phase images have a resolution four times smaller than the original image in order to correctly sample the fringes in the detector plane. In this paper, we use commercial high-resolution QWLSI (Phasics, Saint-Aubin, France). It provides a lateral sampling of intensity and OPD of 4 × 4 pixels or 29.6 μm × 29.6 μm with 300 × 400 measurement points in the final OPD image. The OPD measurement sensitivity is less than 1 nm [17]. As already mentioned, QWLSI allows wavefront sensing with polychromatic incoherent light, so it is usable with laser beams or halogen white lamps. Both were used in this work.

The physical parameter that is measured with this system is the optical path difference (OPD) of the sample, i.e., $\Delta n \times t$ where $\Delta n$ is the difference in the refractive indices of the sample and the reference medium, and $t$ is the thickness of the sample. In this paper, the OPD that we get from the measurements is a combination of optical index changes and surface height modifications between the reference and irradiated states. In the particular case of laser ablation of a sample of refractive index $n_1$, the refractive index difference between the states before and after the laser shot is typically $\Delta n = n_3 - 1$; the thickness $t$ of the ablated area can thus be deduced by the formula $t = \text{OPD}/(n_3 - 1)$ in transmission mode or $t = \text{OPD}/2\times(n_3 - 1)$ in reflection mode. The factor “2” takes into account the fact that the illumination beam has passed in the modified area two times. This simple processing of the data is applied to the measurements that are reported in the paper.
**B. Integration of the Wavefront Sensor on a Subpicosecond LIDT Measurement System**

The first laser experiment used in this study is based on a commercial diode-pumped Ytterbium amplified laser (Amplitude Systèmes S-Pulse HP) centered at 1030 nm, with the possibility of second- and third-harmonic generation (SHG/THG) to obtain 515 and 343 nm wavelengths and sub-500 fs to 3 ps pulse durations (FWHM). The system is described in detail in Refs. [22] and [23]. In these experiments, the laser beam was focused on the entrance surface of the tested sample through a plano-convex lens with an antireflection coating. Polarizers and half-wave plates are used to control the laser fluence incident on the sample. We refer to Ref. [22] for the description of the calibration procedure and measurement of beam parameters (pulse duration and spatial profile). The experimental conditions that have been set for laser irradiation are summarized in Table 1.

The laser beam was directed on the sample with a 45° angle of incidence. An optical microscope (Olympus, BX51) was used for observation of the irradiated area with a x50 magnification long working distance objective (50× Mitutoyo Plan Apo Infinity-Corrected Long WD Objective). The wavefront sensor has been plugged onto the C-mount port of the optical microscope, with the precaution of inserting dielectric filters in the microscope to block the light from the laser. The white light source of the microscope was used for illumination in the experiments, with the wavefront sensor being achromatic. This system was used to observe the irradiated area, facing the sample in normal incidence. A description of this experimental configuration can be found in Fig. 2.

The in-line measurement protocol through the wavefront sensor detection is the following. A reference interferogram is first defined just before the laser shot, as the average from 30 successive interferograms measurements. Then a second measurement of the area of interest is acquired after the laser shot, from the average of 1 to 20 successive interferogram acquisitions (depending on the OPD distribution range of the laser damage). Each averaged interferogram (before and after the damage test) is processed in order to recover both the intensity image of the sample and its OPD distribution. Using such a protocol, the measurement is free from any perturbation that can be introduced by the imaging system, and the processing time is less than 150 ms for processing full frame images. Typically in these experiments, image acquisition takes 50 ms (exposure time setting), and the processing time is less than 150 ms for full frame images. The total time duration for a complete measurement (reference, laser shot, interferogram acquisition, and processing) was less than 2 s in these experiments and could be easily reduced.

**Table 1. Laser Irradiation Parameters Used in the Experiments for the Subpicosecond Irradiations**

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Pulse Duration (FWHM) (fs)</th>
<th>Beam Diameter in Sample Plane (Normal Incidence) (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1030</td>
<td>560</td>
<td>75</td>
</tr>
<tr>
<td>343</td>
<td>350†</td>
<td>35</td>
</tr>
</tbody>
</table>

†The pulse duration for these experiments have been measured at 1030 nm.

**C. Integration of the Wavefront Sensor on the CO2 Laser Processing Experiment**

In order to test the potentialities of the wavefront sensor to measure deep and large laser-created craters, we have implemented the system on an experimental bench that is used for local laser processing of fused silica samples. The principle of operation of the bench is described in Ref [24]. Basically, a CO2 laser beam is directed and focused on the surface of a silica sample. Due to the high absorption factor of the silica at 10.6 μm (85%), the silica is efficiently heated and a crater is formed under the irradiated area by material vaporization. The crater depth and lateral dimensions depend on the beam size, the incident power, and the duration of the irradiation. Typically in this work, we have made craters from few microns to more than a hundred microns, with millimeter size diameters.

The wavefront sensor has been implemented on a microscope (same model as the one described previously) for observation of the irradiated area. The sample was mounted on a translation stage and the following protocol has been used: acquisition of a reference under the microscope, displacement of the sample to the laser irradiation location, laser irradiation, displacement of the sample to come back under the microscope, and acquisition of the measurement of interest. For these experiments, we have used a red diode laser collimated beam for the illumination, which was done in transmission mode. The configuration is shown in Fig. 3.

**D. Samples**

Two different kinds of samples were investigated in our study: fused silica and optical coatings. The optical properties of these...
samples, and in particular the refractive index \( n \), are of main importance because the optical thickness is the parameter that is measured with QWLSI. The value of \( n \) is therefore required if one wants to extract the thickness from the OPD measurement.

The fused silica samples used were made of Corning 7980 material. The samples were 25 or 50 mm in diameter and polished by THALES-SESO. Optical properties of these samples are well known and were extracted from handbooks.

Single-layer thin films of HfO\(_2\) and Nb\(_2\)O\(_5\) deposited on these fused silica substrates were also under investigation. The HfO\(_2\) and Nb\(_2\)O\(_5\) samples were deposited by magnetron sputtering (MS). Specifically, the “Hafnia” samples contain 5% silica. Another Nb\(_2\)O\(_5\) was also deposited by an ion-assisted deposition (IAD) process. The optical properties were determined from spectrophotometry measurements of reflection and transmission and an appropriate fitting algorithm [25]. Table 2 shows the determined parameters of the different samples.

### E. Off-Line Characterization Systems for Comparative Measurements

We have used different commercial characterization systems in this study for comparative measurements of quantitative phase data or to have a deeper knowledge on the observed laser-irradiated sites. They are

- differential interference contrast (DIC) optical microscope: Zeiss Axiotech with \( \times 20, \times 50, \) and \( \times 100 \) magnification objectives;
- scanning electron microscope (SEM): Hitachi TM-1000;
- atomic force microscopy (AFM): Brucker, Dimension-Edge; and
- optical surface profiler (OSP): Zygo Newview.

This last system was used for comparison of quantitative measurements. The optical configuration has been calibrated with a reference plane in silicon carbide to correct residual optical aberration. The axial resolution is about 1 nm and the lateral resolution, defined by the camera pixel, is equal to 1.1 \( \mu \)m.

### 3. RESULTS AND DISCUSSION

#### A. Quantitative Phase Measurements on Laser-Created Craters on Fused Silica

In order to evaluate the accuracy, the dynamics, and the limitations of the system in terms of OPD measurements, we have chosen to study laser-irradiated sites on fused silica samples. On one hand, fused silica is an optical material with well-known optical properties. On the other hand, this material can be easily laser processed to obtain very smooth and clean craters in a large range of depths. Experiments have therefore been conducted with the femtosecond laser system to obtain craters in the nanometer range to explore the low OPD case and with the CO\(_2\) laser system to obtain craters in the hundreds of micrometers range to explore the high OPD case.

We show in Fig. 4 measurements obtained with the wavefront sensor and the associated OSP and DIC measurements of the same sites on the silica sample irradiated at 1030 nm, 500 fs, with single pulses between 3.3 and 5.3 J/cm\(^2\) and with an incident angle of 45°.

A comparison of the measured depths and profiles obtained with the wavefront sensor and the optical surface profiler is shown in Fig. 5. The depth is defined as the deeper measurement compared to the surface level outside the laser-irradiated area.

If we compare the results at high fluence, the agreement between the two techniques is correct. This is particularly evidenced with the profile for the 5.24 J/cm\(^2\) site given in Fig. 5. However, at low fluences, there are differences between the results of the two measurements. One of the possible reasons is that at low fluence, the damaged sites are not smooth anymore: the sites exhibit high roughness and some pits that are not resolved equally with the two systems. This is particularly evidenced in Fig. 5. Additionally, when the depth is around 1 nm, the limit of the system is reached.

We show in Fig. 6 the case of the CO\(_2\) craters. The irradiation conditions are given in the legend of the figure.

![Fig. 4. Surface topography maps obtained with the in-line wavefront sensor (left column) and the optical surface profiler (middle column) on laser-irradiated sites on the silica sample. Test conditions: 1030 nm, incident angle of 45°, fluence of 3.78 J/cm\(^2\) (first row), 4.33 J/cm\(^2\) (middle row), and 5.24 J/cm\(^2\) (last row). Each line corresponds to the same sites observed by the different techniques. DIC microscopy images are also reported in the right column. The length of the scale bar is 10 \( \mu \)m.](image)
A comparison of the measured depths and profiles obtained with the wavefront sensor and the optical surface profiler is shown in Fig. 7. Again, the depth is defined as the deeper measurement compared to the surface level outside the laser-irradiated area.

An excellent agreement is obtained for all the measured sites. Other experiments were conducted for different irradiation parameters (larger beam size, for example) and the results are the same in the range of 1 to 100 μm that has been investigated. Larger and deeper sites were not investigated because of the limitation of the field of view with the available microscope and objectives. We can note, as illustrated in the figure, that the agreement is very good for the measurements on the rim profiles around the craters (these rims are formed by the material’s viscous flow during the crater formation). As also illustrated in the figure, the depth is measured accurately with the two systems. However, the complete profile cannot be obtained with the optical surface profiler, possibly because the flanks of the crater are too steep. The complete profile is obtained with QWLSI.

**B. Laser-Induced Damage Threshold Measurements with Phase Imaging**

As discussed in Section 1, the damage detection system is a key element in any LIDT experiment. The investigated technique in our present work is of interest in several applications: for high sensitivity and spatial resolution, it can be applied in reflection or transmission mode, and it can be implemented in an automated system using simple image processing algorithms [8]. We have therefore compared the LIDT results obtained with QWLSI with the DIC detection method, which is a recommended method by the ISO standard. Damage tests were done on the samples in Son1 mode (1, 10, 100, or 1000 laser shots per site), with 10 tested sites per fluence. The detection through QWLSI has been done in-line, i.e., with a direct
comparison of the surface state before/after a shot on the sub-picosecond experiment described before. After the tests, the DIC microscope has been used for off-line observations of the tested sites. Damage in this case was determined by comparing the investigated site with the surrounding area. Damage was considered as any modification detected through the concerned detection method. We report on these results in Fig. 8. The LIDT is given as the incident fluence on the samples, which were irradiated at 1030 nm, 45° of incidence, and P polarization.

LIDT measured with the two techniques are very close to each other. An interesting result, however, is that for most of the samples, there are some modifications that can be quantified by QWLSI below the limit of detection with DIC microscopy, particularly for 1 or 10 pulses, because for a larger number of pulses the damage is quite catastrophic and easy to detect. In the case of silica for instance, the difference is above 10% for single pulses. This is illustrated in Fig. 8(b) where the measured OPD is plotted as function of fluence and compared to the DIC probability curve. Similar observations can be made with the Niobia sample [Fig. 8(c)] or the Hafnia samples. These small modifications detected by QWLSI could be due to some refractive index changes, mechanical modifications, or a combination of both. There are some fundamental reasons for this observation. Indeed with DIC, an image corresponding to the phase gradient in one direction is obtained, with this gradient being related to the prism that is provided by the microscope manufacturer. DIC microscopes are mainly optimized for biological applications with large OPD distributions in the images (typically 100 nm). QWLSI has been designed for reaching a few nm in the OPD measurement. Additionally, even if the technique is also based on the acquisition of the phase gradients, it records such values in two directions, allowing a complete phase retrieval process. Therefore, we propose to analyze in the next section the quantitative information provided by QWLSI for the analysis of the damage process.

C. Analysis of a Femtosecond Laser Damage Process with Phase Imaging

Interest in the quantitative information that can be acquired during laser interaction experiments is illustrated and discussed in this section through the analysis of an example. The studied

![Fig. 8](image)

**Fig. 8.** (a) Comparison of LIDT measurements with detection done in-line with the wavefront sensor or off-line with the DIC microscope. LIDT values are reported for 1030 nm, 45° of incidence, P polarization, 560 fs. “Difference in %” on the right vertical axis quantifies the difference between DIC and QWLSI LIDT. Two complete measurements are also reported with indication of the LIDT (orange circle): (b) one-on-one measurements on the silica sample at 1030 nm and (c) one-on-one measurements on a Niobia sample at 343 nm.

![Fig. 9](image)

**Fig. 9.** Observation with the wavefront sensor of laser-irradiated sites on Niobia film. Test conditions: 343 nm, 350 fs, single shot, normal incidence. The left columns are the obtained measurements, and the right columns are profiles of the OPD measurements taken in the center of the image in the horizontal direction. Each line corresponds to a different site, with the corresponding laser fluence indicated. The length of the scale bar is 10 μm.
sample is a coating of Nb_2O_5 deposited on a fused silica substrate with the IAD technique (see Table 2). Different sites on this sample were exposed to subpicosecond, 343 nm, single-shot pulses, in normal incidence. Other conditions were described above and were the same for this experiment. In Fig. 9, we report on some of the quantitative phase measurements that were obtained at different fluence levels.

In this particular sample, we observe a damage process that occurs in different steps: first by some surface modifications and second by a delimitation of the film. The fluence threshold for these two steps with QWLSI measurements are 0.15 J/cm^2 for the surface modification and 0.26 J/cm^2 for the delamination threshold. These two steps are also observed with a DIC microscope and the respective measured thresholds are 0.18 and 0.26 J/cm^2. The difference in LIDT between the two techniques comes from the fact that, as shown in Fig. 9, the surface modification at low fluence can correspond to less than 1 nm OPD which is not observed with the DIC microscope.

The first step of the damage process corresponds to the formation of a “bump” under the irradiated surface. This formation of a bump has been confirmed by atomic force microscope observations, as reported in Fig. 10. However, if the diameter of this bump is found the same as with AFM and QWLSI measurements, the measured height is different when we compare the two techniques for all the investigated sites. In the case of AFM, only topological variations are measured. However, in the case of QWLSI measurements, both the refractive index and the surface height contribute to the phase variation. Therefore, this difference may be attributed to refractive index variations (a modification of the refractive index of 10^{-2} on a length of 100 nm can result in an OPD variation of 1 nm). Such modifications of the film possibly result from thermal effects due to the rapid heating and cooling of the film after laser irradiation.

When the fluence is increased up to 0.25 J/cm^2, a strong increase in the OPD value occurs at the center of the irradiated area (Fig. 9). In order to investigate the possible cause, we made an AFM measurement of this area, as reported in Fig. 11. This measurement shows the formation of large material grains that could be related to the high temperature reached in this area and possibly crystallization of the film. Since the structure is submicronic, it cannot be resolved with QWLSI and a mean value of OPD is measured in this area.

At higher fluences (>0.25 J/cm^2), part of the film is removed, as shown in Fig. 9. The higher the fluence the larger the diameter of the removed film area, but the depth stays quite the same. Comparison between AFM and QWLSI measurement shows that the two techniques give the same depth value of around 40 nm. Simulations of the electric field distribution in the film under the conditions of laser irradiation indicate a maximum of the electric field located at a depth of 44 nm in the film (Fig. 12). This value matches the experimentally measured depth of the damage area, indicating that damage initiation occurred in that location.
Following this experiment, it appears that the quantitative phase information that can be obtained from the measurement system is well suited for laser interaction studies: different stages of laser material modifications can be evidenced with high sensitivity corresponding to OPD variations of less than 0.5 nm.

4. CONCLUSIONS

As a conclusion, we have shown that in-line quantitative phase microscopy is a very valuable tool for LIDT metrology purposes and to study damage processes. It allows for precise damage detection and can give better sensitivity for LIDT measurements compared to classical microscopy detection systems. Additionally, quantitative information can be obtained on the laser damage sites, with an optical path difference better than 0.5 nm and the three-dimensional characteristics of laser damage or laser ablation zones can be determined accurately. The main interest for this application is that the technique can be implemented in a simple macro or microscope and the measurement is immediate.

These points are particularly useful in the study of laser material interactions and are of main interest in the understanding, for instance, of the physical process involved. Eventually, using pump–probe techniques adapted to optical microscopy, it should be possible to obtain time-resolved intensity and phase information that can be obtained from the measurement system is well suited for laser interaction studies: different stages of laser material modifications can be evidenced with high sensitivity corresponding to OPD variations of less than 0.5 nm.

REFERENCES