Time-resolved quantitative-phase microscopy of laser-material interactions using a wavefront sensor

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Received 10 May 2016; revised 16 June 2016; accepted 17 June 2016; posted 17 June 2016 (Doc. ID 264906); published 8 July 2016

We report on a simple and efficient technique based on a wavefront sensor to obtain time-resolved amplitude and phase images of laser-material interactions. The main interest of the technique is to obtain quantitative self-calibrated phase measurements in one shot at the femtosecond timescale, with high spatial resolution. The technique is used for direct observation and quantitative measurement of the Kerr effect in a fused silica substrate and free electron generation by photo-ionization processes in an optical coating. © 2016 Optical Society of America

OCIS codes: (140.3330) Laser damage; (190.3270) Kerr effect; (320.7100) Ultrafast measurements.

http://dx.doi.org/10.1364/OE.41.003245

Laser-material interactions at high intensity have important implications for many scientific applications. To understand the physical processes that occur in such interactions, for instance, material excitation and response to laser irradiation, experimental tools that allow observation and analysis of in situ and in real time of these interactions are valuable. For this purpose a large variety of so-called pump/probe techniques exist based on monitoring the change of properties of a probe beam (reflection, transmission, phase shift, etc.), passing in an area where a pump beam induces the excitation. With the use of very short laser pulses, a high temporal resolution can be obtained on the material response dynamics. Such techniques in a microscope configuration are also possible to obtain information on the excited zone with high spatial resolution. Studying the spatio-temporal dynamics of the laser-material interaction process can be of much interest for fundamental purposes (carriers or plasma dynamics) or applications (laser processing of materials, laser damage of optical components). However, to obtain a complete picture of the material optical properties at a given time, for instance, the real and complex parts of the refractive index, it is necessary to simultaneously acquire amplitude and phase images of the excited zone. For that purpose several techniques have been proposed. Without being exhaustive, they include phase contrast microscopy [1], spectral interferometry [2,3], digital holography [4–6], and phase quantitative non-interferometric technique [7]. Even if very suitable for the technique’s purpose, each has drawbacks: phase contrast microscopy does not allow quantitative measurements; spectral interferometry is intrinsically limited to 1D spatial resolution, the second dimension on the sensor being used for the spectral information; and digital holography is complex to implement and requires specific algorithms for the phase retrieval. In this Letter we propose an alternative technique to obtain time-resolved intensity and phase images of an ultrafast event based on direct wavefront analysis of a probe beam. It is shown that the technique is straightforward (self-calibrated) and highly sensitive, and presents a diffraction-limited spatial resolution. It is applied to study the nonlinear Kerr effect in a fused silica glass and picosecond laser damage process in an optical coating.

The wavefront sensor used in this work is a commercial system (SID4Bio, Phasics, Saint-Aubin, France) based on Quadriwave Lateral Shearing Interferometry (QLSI) [8]. Briefly, a diffractive optics element makes four replica of the incoming beam to form an interference pattern that is recorded on a CCD sensor. Because the wavefront sensor is placed in the image plane of the microscope, such an interferogram therefore contains two-dimensional information relative to the Optical Path Difference (OPD) gradients in the image plane [9]. Integration of these gradients makes it possible to recover the actual OPD distribution of the imaged sample. In Ref. [10] we have shown that by coupling this wavefront sensor with a high-magnification optical microscope operating in white light illumination, quantitative-phase information with an OPD better than 0.5 nm could be obtained and the three-dimensional characteristics of laser damage or ablation zones could be determined accurately. In the present work we now investigate the possibility of using such a system to obtain time-resolved intensity and phase information to study the dynamics of the laser-material interaction process. This implies that the measurement should be done in one shot, with a coherent source that introduces speckle noise and with pointing instabilities that degrade the referencing procedure. The developed experiment for this study is described in Fig. 1.

The laser source was operating at 1030 nm with 1.0 ± 0.1-ps pulse duration. The pump beam was focused on the sample with a beam diameter of 50 ± 2 μm at 1/e².
It has a close-to-Gaussian profile and was incident on the sample with an angle of 51°. The fluence incident on the sample is given in this Letter as in the perpendicular plane of the beam. As described on the figure, the probe beam (515 nm) was the second harmonic of part of the pump one. Once spatially filtered, its diameter (around 1-mm diameter for the present experiments) was adapted to largely cover the imaging field of view given by different long-working-distance objectives (50 ×, 20 ×, and 10 × APO infinity-corrected, Olympus, Japan) used in our optical microscope (BXFM, Olympus, Japan). Probe illumination was used as the light source in the microscope working in transmission mode. The probe energy was kept below 1 μJ, and its fluence on the sample was negligible compared to that of the pump one, implying no contribution to the excitation process. The wavefront sensor has been plugged on the C-mount imaging port of the optical microscope. Filters were inserted between the objective and the tube lens to block the pump beam scattering (KG3, Schott, Germany) and minimize the contribution from the broadband thermal radiation from the plasma in case such event occurs (FL514.5-10, Thorlabs, Germany). The integration time on the sensor was reduced to 1 μs to minimize the noise. The delay between pump and probe was adjusted with a motorized translation of retroreflectors along the pump beam path. The pump beam diameter variation due to such a change in the pump path length was found to be less than 5% in the sample plane. Pump and probe were linearly polarized with S polarization in the sample plane.

The time-resolved measurement protocol through the wavefront sensor detection was the following:

1. Acquisition of a reference averaged interferogram from 30 successive probe illuminations, without pump excitation.
2. Acquisition of one interferogram obtained from a single-probe pulse illumination, in combination with a controlled delayed pump excitation pulse in the field of view.
3. These two interferograms are processed so that only changes in the optical parameters of the sample due to the pump pulse are accounted for in the resulting OPD map.

The system has been first used to study the OPD changes caused by the nonlinear Kerr effect in a fused silica round plate (C7980, Corning USA), 50-mm diameter, 5-mm thick, super-polished for high-power laser applications (Thales SESO, France). Indeed the Kerr effect is well known and the nonlinear refractive index \( n_2 \) of fused silica is well documented [11]. This case can thus be considered as a reference for validation of the technique. The pump beam was incident as described previously, and a damage threshold of \( 10.5 \pm 0.5 \text{J/cm}^2 \) was measured at 1030-nm, 1-ps, single-pulse irradiation, 51° AOD. Considering the angle of incidence and pulse duration, this value is in accordance with previous measurements on the same system and calibration procedure at 500 fs [12] (comparison is made using the temporal scaling law described by Mero et al. [13] and electric field distribution scaling as described in Ref. [12]). This is an important point since the absolute fluence value is required to compare experiments to simulations. Under such conditions we have recorded the time-resolved OPD maps for different fluences (from 6 to 10 J/cm² with \( \approx 0.5 \text{J/cm}^2 \) increments) and different pump/probe delays (0 to 6 ps with 0.33 ps increments). Five measurements were taken for each parameter (fluence, delay), and in the following we provide the mean value. Typical OPD maps recorded at different delays are reported in Fig. 2. For these measurements the microscope was focused on the entrance face \((z = 0)\), and the OPD variation related to the propagation of the pump beam in the sample can be clearly observed. The 6-ps timescale was limited by the field of view. Figure 3 gives the evolution of both maximum and profile along the vertical axis of recorded OPD distributions (see Fig. 2), as a function of fluence and/or delay.

To interpret these results we have used a simple numerical model. It is based on the calculation at a given location in the sample of the refractive index modifications, accounting for the temporal pulse profiles of pump and probe, their respective delays, and the spatial distribution of the pump (probe is considered homogeneous). The refractive index variation is then integrated along the probe beam path and time integrated.

The OPD measured in the sensor plane can then be written:

\[
\text{OPD}(x, y, D) = T \int_0^{\infty} \int_{-\infty}^{\infty} n_2 I_p(t, x, y, z) I_0^N(t- D, x, y, z) dz \, dt
\]

with \( D \) the temporal delay between pump and probe, \( T = 1 - R \) the coefficient of transmission through the first interface, \( \epsilon \) the sample thickness, \( n_2 \) the non-linear refractive index, \( I_p \) the pump intensity with \( I_p(x, y, z, t) = I_0^p \text{sech}^2\left(\frac{t-nz/c}{\text{tan} \theta_z}\right) \text{exp}(-2 \frac{\text{tan} \theta_z}{\text{tan} \theta_0} \frac{z}{\omega_0^2}) \text{exp}(-2|\text{tan} \theta_z|/\omega_0^2) \), \( I_0^N \) the normalized probe intensity with \( I_0 = I_0^p \text{sech}^2\left(\frac{t+nz/c}{\text{tan} \theta_0}\right) \), \( n \) the refractive index, \( \theta_0 \) the angle of incidence in the material, and \( \tau_p \) and \( \tau_r \) the pump and probe pulse duration, respectively. For the simulations the input parameters are the beam size, pulse duration, refractive index, angle of incidence, sample thickness, and fluence, all of which are known or have been measured. Only the zero-delay was adjusted so that experiments and simulations correspond to the same time base. For \( n_2 \) we have used the value 3 cm²/W [14,15] for the simulations. Comparisons of experimental data to numerical simulations reveal an excellent agreement (Figs. 2 and 3) for quantitative OPD value and its spatio-temporal evolution. This demonstrates that accurate OPD maps can be obtained with high spatial resolution (given by the effective...
OPD pixel size of 30 μm divided by optical magnification \[10\] and temporal resolution (limited by the probe pulse duration). It must be also noticed that the processing time for OPD reconstruction is less than 150 ms for a full frame image on a laptop computer, allowing real-time observations.

As a second application of the system, we have studied a Nb$_2$O$_5$ film deposited on a fused silica substrate. The film has a thickness of 231 nm with a refractive index of 2.21 at 1030 nm. The sample was studied in similar conditions as described previously, excepting that the angle of incidence was 45° and the pulse duration 1.5 ps, and we used an objective with higher magnification (×50). On this sample the pump fluence was above the Laser-Induced Damage Threshold (LIDT) to observe the material excitation and response leading to damage of the film. Nb$_2$O$_5$ has a low LIDT compared to silica so the damage process in the film at highfluence can be studied without damaging the substrate.

**Fig. 2.** OPD measurements corresponding to phase shifts induced by the pump beam (1030 nm, 1 ps, J/cm$^2$) on the probe beam (515 nm, 1 ps) propagating in a fused silica sample. At time $t = +0.7$ ps the pump beam is entering the sample; at time $t = +1.3$ ps the main part of the pump is in the sample; and for higher delays the pump is located in the bulk of the sample and propagates toward the exit surface (see inset of Fig. 1). On the right-hand side we report on the simulations of the expected OPD distribution based on the experimental configuration and $n_2 = 3.0 \times \text{cm}^2/\text{W}$. Experiments and simulations have the same scale in $x$, $y$ and OPD.

The main interest of our technique is allows simultaneous recording of amplitude and phase information, as reported in Fig. 4. In this figure, OPD images were obtained with the same procedure as previously described, and amplitude ones are given as transmission ratio: the amplitude image of interest was divided by the amplitude reference one to obtain the changes in transmission.

If we look first at the transmission ratio images, the measurements reveal a strong drop in transmission of the laser irradiated area. This can be classically linked to the increase of free electron density in the materials due to ionization processes [6,16], followed by a partial transmission recovery related to the decrease of the free electron density. However, when the pump pulse is in the field of view of the imaging system we notice a strong increase in transmission. This effect is not well understood, but we attribute it as an artifact linked to the Kerr effect in the substrate. Indeed as shown on the associated OPD maps, the Kerr effect in the substrate is clearly seen as in the previously reported results. The increase of free electron density generates a drop in transmission, related to the increase of the extinction coefficient, but also a strong phase shift related to the modification of the real part of the refractive index [17]. This can be clearly observed. The simulation of these effects requires complex simulations and is out of the scope of this Letter. However, to emphasize the interest of the technique we report in Fig. 5 transverse profiles of OPD and transmission ratio maps at a particular delay. One can clearly observe in this figure a saturation of the transmission ratio (flat top profile) and a fluence-dependent distribution of the OPD

![Fig. 3.](image-url)
These measurements give evidence of a saturation of the extinction coefficient at a given fluence and a dependence of the real part of the refractive index with fluence. Interpretation of such changes in the sample optical properties with an appropriate model should give meaningful information about electronic density with respect to irradiation conditions.

To conclude we have shown that quantitative time-resolved phase microscopy is a valuable tool for the study of transient laser-material interactions. Because of the interesting characteristics of the wavefront sensor (sensitivity, self calibrated, robustness) and the high spatial and temporal resolution of the proposed technique, many applications seem to be reachable. As examples, single-shot measurements of non-linear properties of transparent materials should be obtained. Dynamics of laser damage/ablation processes could also be studied since information on the physical processes should be recovered due to simultaneous measurements of transmission ratio and OPD. Moreover, flexibility allows modifying excitation geometry to optimize and thus study specific phenomena (free electron plasma excitation, trapping of defects, phase changes, shock wave generation, material ejection, etc.). Additionally, since the interferogram formation on the sensor does not depend on the wavelength [8], other wavelengths in the sensor range (350–1100 nm) or very short pulses with broadband spectrum are suitable for probe illumination [18].

Funding. Fond pour la Science of the Institut Fresnel.

Acknowledgment. The authors would like to acknowledge Andrius Melninkaitis, Vilnius University, for his advice on the experiment.

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