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Towards in situ monitoring and feedback control of femtosecond laser-induced nanogratings formation in dielectrics

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Abstract

Tightly focused non-ablative femtosecond laser pulses induce a variety of structural modifications in the bulk of dielectrics. Among those, sub-wavelength nanogratings are particularly interesting as a means not only to locally enhance the material etching selectively (and thus, enabling bulk 3D-micro-fabrication), but also for encoding rich information in high-density permanent data storage media. Femtosecond laser-based processes are subject to perturbations, affecting the repeatability and accuracy of the results. To increase the performance of these processes, we explore a feedback method based on direct monitoring of the laser-affected zone (LAZ) using a probe beam. Specifically, we report on the use of weak signals resulting from the interaction of a femtosecond laser probe-beam with the nanogratings index-modulation as objective functions in feedback loop algorithms.

Keywords: femtosecond lasers, laser-induced modifications, dielectrics, feedback control

1. Introduction and problem

When strongly focused inside dielectrics, ultrashort laser pulses trigger nonlinear absorption events, which induce localized modifications of the material structure. These structural modifications are of particular

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interest for applications, such as waveguide writing (Du *et al.* 1994) or selective etching for fabrication (Kondo *et al.* 1999, Marcinkevičius *et al.* 2001, Bellouard *et al.* 2004), among others. The laser-induced modifications can have different morphologies depending on the laser parameters (e. g., wavelength, pulse duration, and energy), processing parameters (e. g., confocal parameters, and exposure dose), and on the type of material being exposed. In the bulk of transparent materials, common modifications include localized densification effects, crystallization and elemental redistribution, or formation of periodic self-organized nano-planes (Shimotsuma *et al.* 2003, Hnatovsky *et al.* 2005).

Like many manufacturing processes, laser-based ones are prone to perturbations, in particular when the foreseen modifications are comparable in size to the laser wavelength. As most of these laser processes are currently performed in open loop, they lack resilience and require prior calibration steps to perform accurately the manufacturing tasks. So far, direct monitoring and thus, control, have with a few exceptions (Hecker, Scharun, and Graf 2021) never been implemented.

Physical processes that lead to bulk structural changes happen both, at ultrafast timescales and at micro to sub-micrometric scales, making their monitoring and control particularly challenging. As most modifications are about the size of the diffraction limit of visible light, classical microscopy techniques unfortunately cannot return high quality images, without using constraining imaging techniques (e. g., very high magnification, and phase-contrast) or non-linear imaging processes.

Femtosecond laser processes involve strong optical intensities (typically > 1 TW/cm^2) that can be used to trigger nonlinear events, such as third-harmonic generation (THG) microscopy, enhanced at interfaces between different index zones. This emitted light offers an efficient signal to visualise localized defects and material phase changes induced during processing, and contains direct information related to structural changes in the material. Pioneered work in this field has been performed by Müller *et al.*, where they reported the use of THG microscopy to image a laser-written waveguide written in fused silica (Müller *et al.* 1998, Schaffer *et al.* 2002). A comprehensive review of THG and more generally multiphoton microscopy is proposed by M. Young and J. Squier (Young *et al.* 2015). However, as THG is very inefficient, the light emitted has a very low intensity and requires confocal techniques with long acquisition times, that makes the method currently unsuitable to implement for efficient process monitoring.

In this work, we use advances in CMOS image sensors to expand the capabilities of THG microscopy to wide-field imaging, in the context of process monitoring, and therefore not just the laser-affected zone, but also its immediate surrounding. Here, we demonstrate the capability to generate full images, wide-field THG images of the laser-affected zones with a high signal-to-noise ratio, in a 10 ms-timescale. Such an advancement could ultimately allow for direct optical observation of the laser-induced periodic structures, given the lowered diffraction limit of the third harmonic (343 nm).

2. Experimental setup

As both machining and imaging beams require pulses in the femtosecond regime, a pump-probe optical layout has been chosen for simplicity. The ability to tune the delay between both beams offers additional flexibility for further time-resolved studies.

In our experiments, the pump beam, which has comparatively the highest focusing power (pulse energy is typically ~ 200 nJ, i.e., 27 TW/cm^2), triggers the cascade of events leading to a permanent structural modification localized in the bulk of the material. On the other end, the probe beam needs to be strong enough to trigger a nonlinear response from the modified areas, yet without inducing any modification into the material.

For triggering the THG response in a volume, large enough to fill the imaging field of view, the probe beam diameter is made smaller than the focusing objective entrance pupil, thus achieving a larger beam diameter on focus than the pump beam. Image formation is finally performed by collecting the transmitted light through a regular microscope objective, and by filtering out everything but the third-harmonic spectral line. The experimental setup is summarised in Fig. 1.

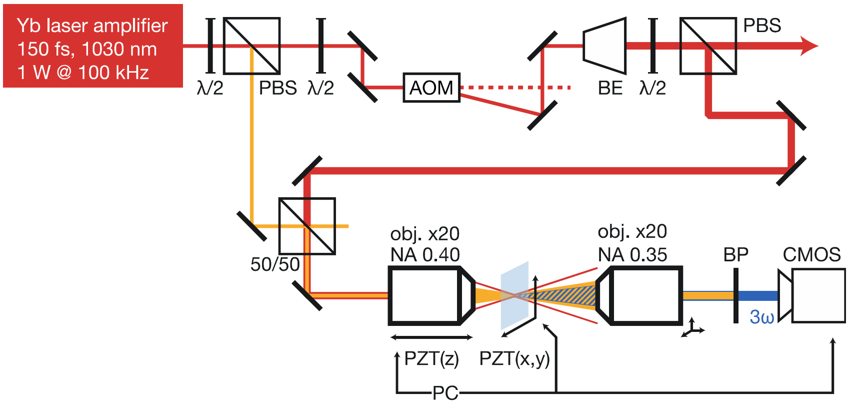

Fig. 1. Diagram of the processing platform with the in-line THG microscope. The femtosecond source is an Yb-based regenerative amplifier (S-Pulse HR SP, Amplitude Systèmes SA), operating at a pulse picking repetition rate of 100 kHz, and at a 1 W of average output power. The pulses are 150 fs long and centred at 1030 nm. The beam is split into a pump (or machining) beam and a probe beam, using a rotating half-wave plate and a polarised beam splitter. The energy of the pump beam is modulated using an acousto-optic modulator. To achieve maximum focusing power, its diameter is tuned to fill the machining lens's entrance pupil, using an adjustable beam expander. Both beams are merged using a 50/50 beamsplitter. The beams are colinearly injected into the machining objective lens (20x, NA 0.4). It is fixed on a piezoelectric-actuated stage (Mad City Labs), to allow for a 100 µm travel range of the focal plane. The latter is laying inside a 250 µm thick fused silica sample (Corning 7980 0F), fixed on two-dimensional piezoelectric stages (also Mad City Labs). The beams are collected using a collecting objective lens (20x, NA 0.35). Two laser-line band-pass filters are used to let only the third harmonic through, i.e. 343 nm, before collection by the low-noise imager.

Here, a new type of ultra-low-noise CMOS image sensor developed at EPFL is used (Boukhayma, Peizerat, and Enz 2016, Boukhayma, Caizzone, and Enz 2019). This dedicated sensor is fabricated in a pinned photodiodes (PPD) based CMOS image sensor (CIS) process. It has more than 50 % quantum efficiency at the third-harmonic wavelength, offering a high sensitivity in combination with a deep sub-electron noise at room temperature, leading to an efficient collection of the weak third-harmonic signal, and enabling live imaging with short integration times of THG.

3. Results and discussions

We validated the sensor technology by using first, an averaged pixel-array and second, the CMOS imager described above.

In the first configuration that uses the averaged pixel-array, a confocal configuration was used, using the pump beam, first to process the sample and second, to induce THG for observation using lower pulse energies. In this confocal configuration, the probe beam shown in Fig. 1 is not used.

In practice, a laser-written test pattern was inscribed at different scanning speeds (100 µm/s and 200 µm/s, respectively) and energies (from 190 nJ to 260 nJ as measured before injection in the objective lens). The sensor integration time was set to 15 ms, while 10 ms-long laser burst of 1000 pulses generated the signal. The latter value is used to compute the rate of counts. The scanning step is set to 0.5 µm in both, x and y directions. In this configuration, the pulse energy used to generate the third-harmonic signal is set at 130 nJ (i.e., 18 TW/cm^2), slightly below the modification threshold, found around 160 nJ. Results are reported in Fig. 2.

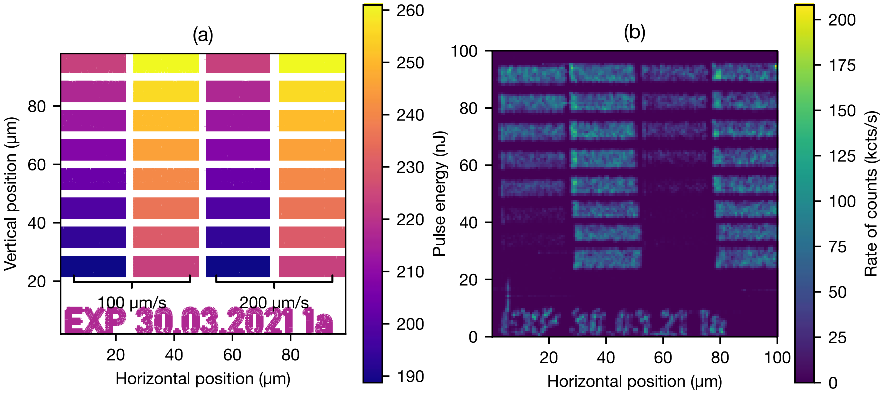

Fig. 2. Inscribed test target (a) and its scanning confocal THG image acquired using an average pixel, ultra-low noise CMOS sensor (b).

The results from the averaged pixel sensor demonstrate the ability of THG to discriminate different levels of material modification exposure dose, especially those obtained at low energy levels (< 220 nJ). By increasing speed, thus lowering the exposure dose, we observe as expected the lowering of the threshold for detecting the modified patterns.

In the second configuration, for wide-field imaging, we use the pump-probe configuration described in Fig. 1. The sensing array is 1.92 x 2.88 mm, while the image is integrated for 38 ms. The probe pulse energy is set to 45 nJ, i.e. circa 2 TW/cm^2 considering a 5.0 µm imaging spot size. Images taken in the vicinity of the right edge of a laser-written line are shown in Fig. 3.

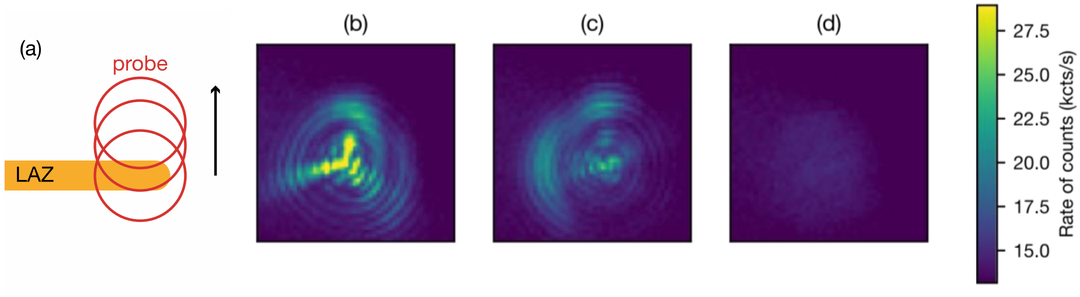

Fig. 3. Examples of wide-field THG images, taken in the vicinity of the right edge of a laser-written line, by moving the specimen across the THG probe signal as shown in (a). Images are taken with the probe being (b) exactly on the edge, (c) 2.5 µm, and (d) 4.5 µm away from it, from the top. Each image is cropped to 50 by 50 pixels. The maximum overall signal is obtained when irradiating directly on top on the laser-written area, and decreases as the irradiation moves away. The overall shape of the laser-written area can be deduced, especially in (b). Observation is however hindered by strong lens ghosting and spherical aberrations, because of the unfitting collection optics.

This is a first proof-of-concept for which the collection optics used here for imaging is not optimized. Furthermore, the actual region of interest covers just a small part of the sensing array and shows strong aberrations. Nonetheless, sharp variations of signal in the THG images depending on the observed region are visible and obtained at rates of up to 30 kcts/s. In Fig. 3(a), the line is itself distinguishable as an enhanced THG source. This exploratory experiment demonstrates the potential of these approaches for observing laser-modified patterns in-situ using THG at a fast rate.

4. Conclusion and outlook

Monitoring laser processes is essential not only for identifying relevant laser process parameters rapidly, but also for implementing closed-loop control algorithms aiming at increasing both, the repeatability and the resolution of these manufacturing processes.

Here, we have reported proof-of-concept results of inline THG microscopy as a monitoring technique of femtosecond laser inscription processes, capable of observing the laser-affected zone proper, but also its immediate surrounding. In practice, a unique low-noise CMOS sensor technology developed at EPFL is used to image *at once* the THG response of a *full observation field* illuminated by a single laser beam. The whole monitoring setup is intrinsically simple and can be mounted in line with a laser processing setup.

As first proof-of-concept, we presented post-mortem images, but the prospect for fast wide-field observations is in sight as it is foreseeable that using this technology, full THG images acquisition at 12 fps capacity during machining achieved on a same experimental setup will be feasible. This would allow for dynamic tuning of the process and ultimately closed-loop control as it would unravel the prospects for higher repeatability and enhanced accuracy of femtosecond laser-based bulk-modification manufacturing.

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In this study, OB, as part of his doctoral work, designed and set the optical setup and the processing platform up. AK characterized and implemented the low noise CMOS array sensor, with the assistance of AB, the original designer of the chip, and AG, under the supervision of CE. Both OB and AK performed the experiments and discussed the results, under the supervision of YB.

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