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Nonlinear Resolution: A Misconception in Femtosecond Laser Ablation

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Abstract: We demonstrate a systematic one-to-one mapping between femtosecond laser ablation features and beam contours at a strict threshold-intensity. This is independent of the nonlinearity of interaction varied using various wavelengths and dielectric materials. ©2020 The Author(s)

1. Introduction

Amplified femtosecond lasers offer the possibility to exceed the threshold of optical breakdown anywhere in the three-dimensional space of dielectrics and semiconductors [1,2], making it a unique and appealing tool for industrial precision machining. The term precision refers, apart from accurate repeatability when using same conditions, to the minimum size of laser induced modifications. On the one hand, sub-diffractive periodic modifications could be obtained after multipulse irradiation, formed by patterned electric-field enhancement due to surface inhomogeneity [3]. On the other hand, if willing to use this technology as material-independent precision tool, progress on digital laser writing resolution should be a priority. For that purpose, the intensity-threshold response of femtosecond laser material ablation was exploited for resolution machining demonstrations by tight focusing, reaching craters as small as $\cong 40$ -nm irradiating at 1053 nm ($< \lambda/20$) [4]. Additionally, as exploited in multiphoton microscopy, it is commonly accepted a reduction of resolution produced due to the confinement of the non-linear absorption profile [5]. In this paper, we undoubtedly demonstrate this nonlinearity-based benefit is actually irrelevant for resolution purposes in femtosecond laser machining. More generally we establish this conclusion holds for any observable relying on an intensity-threshold phenomenon, as the case of femtosecond laser material ablation. By solving this misconception that have concentrated the machining studies in the visible and near-infrared regime, we have a possibility to re-route developments towards practical solutions for direct laser writing at the nanoscale.

2. Experimental results and discussions

To address experimentally the question, we have chosen high-order nonlinear absorption conditions. As illustrated with fig. 1b, we perform single-shot irradiations on sapphire (band gap $E_g=8.8$ eV), with pulses at wavelengths as high as 1550 nm ($E_{ph}=0.8$ eV) and 190-fs pulse duration at FWHM. Thus, the energy of 11 photons is required to cross the band gap. The infrared pulses are delivered by an optical parametric amplifier (OPA) pumped with pulses of 500 μ J at 1030 nm (Pharos, Light Conversion). An aspheric lens ($f=50$ -mm) focuses the beam onto the sample, which is mounted on XYZ motorized stages. Motion in the XY plane allows positioning the sample on a fresh region from pulse to pulse. Micrometer precision Z-axis motion and in-situ surface microscopy imaging (10x, tilted at 45°) ensures precise positioning. Afterwards, craters are characterized by means of confocal microscopy (Leica DCM3D, 460-nm illumination, 150x objective lens), allowing to measure the ablated area with sub-micrometer lateral precision. The demonstration is carried out by direct comparisons between measurements of the spatial characteristics of the produced craters and the beam profiles recorded with an in-situ imaging system.

The excellent agreement obtained between the contours of the beam profile at 1550 nm at threshold fluence F_{th} (shown in figure 1b at $F_{peak}=1.4 \times F_{th}$) and the size of obtained craters shows that the knowledge of the fluence distribution makes possible to retrieve the beam portion exceeding the fluence ablation threshold. In order to generalize this conclusion that contradicts the concept of nonlinear resolution, we have repeated similar experiments in different materials: fused silica suprasil, fused silica infrasil and a soda-lime glass, with respectively optical band gaps of 9 eV, 5.2 eV and 3.8 eV. For even higher change of the nonlinearity order we also preform the experiments at different wavelengths. An example is shown in figure 1a with beam profile and material responses at 515 nm obtained by second harmonic generation.

The direct correlation between fluence profile “thresholding” and the induced craters obtained for all cases is visually expressed with Figure 1. However, more detailed analyses (not shown here) reveal slightly smaller ablated features in soda-lime. Interestingly, it is the lowest band gap material tested in our experiments. This allows to

directly exclude a potential nonlinearity-based confinement of ablation, attributing the size reduction to a specific material response. Given the observation of elevations at the crater edge (see Fig. 1) and the borosilicate nature of soda-lime, we conclude on a similar hydrofluidic phenomena as the one explained by Ben-Yakar *et al.* [6]. At the opposite, sapphire exhibits very neat craters and can be consequently considered as a reference dielectric for demonstration of the threshold nature of femtosecond laser ablation.

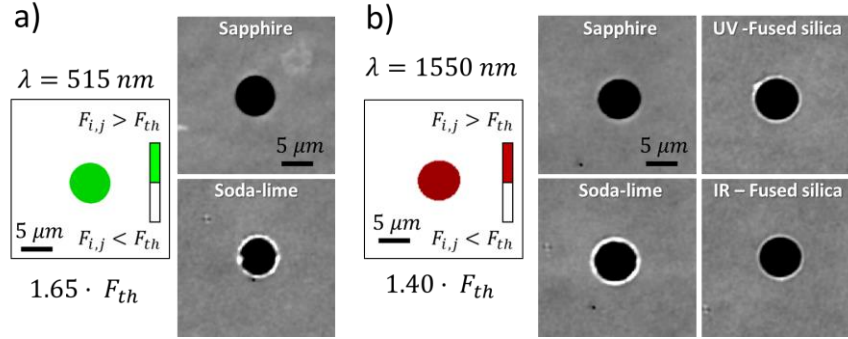


Fig. 1. Comparison between laser profiles and corresponding impacts for different wavelengths and materials (a) At left, beam portion above the ablation fluence threshold a given excitation level ($F_{\text{peak}}/F_{\text{th}}$) for a 515-nm femtosecond laser beam. Pixels having values above the fluence threshold are represented in color and pixels below the threshold in white. At right, confocal microscopy images of the surface modifications on sapphire and soda-lime induced at the indicated excitation level. Surface depressions and elevations are respectively represented with darker and brighter color. (b) Similar than in (a) but for a 1550-nm irradiation. Together with sapphire and soda-lime are shown the results on two different samples of fused silica.

The main conclusion of this study is the absence of nonlinear resolution in all tested femtosecond laser ablation conditions. The only small deviations in the predictions of crater shapes and sizes by pure “thresholding” of the fluence profiles rely on the material-dependant hydro-dynamical response but is independent on the nonlinearity order. Looking at the perspectives after this demonstration, it is striking to observe the opposite directions taken by femtosecond laser machining and lithography that are two fabrication methods on which the same optical limits finally apply. On the lithographic front, technologies today rely on the use of extreme UV illumination and a sub-20 nm resolution is routinely achieved. On the laser front, high-precision machining remains primarily based on ultrafast lasers emitting in the near-infrared. Despite similar resolutions have been demonstrated in laboratories [4], laser machining falls short in providing a practical solution for fabrication at the nanometer scale. There has been probably no technological disruption because the discussed misconception has associated high-resolution to strong nonlinearities (and so long wavelengths). By the introduction of shorter wavelengths, similarly to lithography, this vision could open a direct and simple route to reliable femtosecond laser machining at the nanometer scale.

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