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Pulse-duration dependence of laser-induced modifications inside silicon

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Abstract: The advent of ultrafast infrared lasers provides a unique opportunity for direct fabrication of three-dimensional silicon microdevices. However, strong nonlinearities prevent access to modification regimes in narrow gap materials with the shortest laser pulses. In contrary to surface experiments for which one can always define an energy threshold to initiate modifications, we establish that some other threshold conditions inevitably apply on the pulse duration and the numerical aperture for focusing. In an experiment where we can vary continuously the pulse duration from 4 to 21 ps, we show that a minimum duration of 5.4 ps and a focusing numerical aperture of 0.85 are required to successfully initiate modifications. Below and above thresholds, we investigate the pulse duration dependence of the conditions applied in matter. Despite a modest pulse duration dependence of the energy threshold in the tested range, we found that all pulse durations are not equally performing to achieve highly reproducible modifications. Taken together with previous reports in the femtosecond and nanosecond regimes, this provides important guidelines on the appropriate conditions for internal structuring of silicon.

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1. Introduction

The emergence of ultrafast laser writing in narrow bandgap materials offers the possibility for directly creating microdevices with 3D architectures for application fields as important as micro-electromechanics and silicon photonics. The first recent successes in locally tailoring the properties of silicon (Si) with ultrashort pulses tightly focused below the surface [1–5] have revealed responses strongly deviating from those extensively studied in dielectrics during the last two decades [6,7]. Till now, the investigations for internal modification of silicon have been focused on the short-wave infrared region of the spectrum (SWIR) and very modest dependence to the wavelength parameter is found as long as multiphoton absorption initiates localized energy deposition near the focus in an originally transparent Si target [8,9]. This is very similar to how visible or near-infrared femtosecond pulses are used for 3D writing applications inside dielectrics. However, the dependence of the problem to the pulse duration remains a large matter of debate. The energy threshold for modification of a material normally decreases with pulse shortening because of the delivery of progressively higher peak intensity and so more efficient nonlinear absorption. This is a picture common for any surface machining studies [10,11] and internal dielectric modifications [12]. However, it remains striking to note that opposite responses are reported in bulk-Si interaction studies. In particular, nanosecond pulses are today readily used for internal inscription in silicon leading to technologies as important as wafer stealth dicing [13] or internal marking [14]. It is also today used for more advanced processing with some optical and fluidic functionalities added to Si in this regime [15–20]. At the opposite, the shortest ultrashort pulses (sub-100 fs) usually fail in inducing permanent changes inside Si using conventional machining configurations [2,8,21,22]. One can also note contradictions in the intermediate picosecond regime where some argues that modification regimes are hardly achievable with pulses of few picosecond duration [5,23] while other have recently presented

demonstration of waveguide writing in Si with sub-ps pulses and conditions very similar to those used in dielectric studies [3,4]. As we will show in this paper, the controversy persists because the space-time localization of light inside Si-bulk rely on a stronger interplay between nonlinear processes across the focus than for dielectrics [8,21,24,25]. This leads to important dependences to the various experimental parameters making hard to make comparisons between experiments where the pulse duration is changed. This strong interplay is also causing an increased sensitivity to fluctuations in the experimental conditions. It is the subsequent lack of repeatability in the observations that has likely limited the conclusions of a first attempt in investigating the pulse duration dependence of this problem in the picosecond regime [23]. One aim of this paper is to definitively answer the question of the pulse duration dependence by designing and performing an experiment where the pulse duration is varied by chirping SWIR laser pulses generated by optical parametric amplification. In this way, we deal only with pulses of high temporal contrast, the latter being another critical parameter [26].

The main contributions that prevent modification with the shortest pulses are beam depletion by multiphoton absorption, Kerr-induced phase distortions, screening and defocusing of strong plasma created prior the focus [2,5,8,24]. All these phenomena are intensity-dependent nonlinear effects increasingly limiting the energy density that can be delivered as the pulse duration is decreased. Such limiting factors come directly into picture when the clamping of delivered intensity is measured [2,22]. Another objective of this paper is to access experimentally the pulse duration dependence of the delivered conditions so that we can extrapolate from measurements below material breakdown condition, the laser technologies and temporal regime which are the most inclined to cross the threshold for internal Si writing. These must serve as some important guidelines for future developments in the challenging topic of high-precision 3D ultrafast laser writing in silicon.

2. Experimental setup

The experimental setup is shown in Fig. 1(a). The femtosecond laser source (Pharos, Light-conversion) delivers pulses at 1030 nm central wavelength. The wavelength is then converted to 1550 nm by using an optical parametric amplifier (OPA, Orpheus, Light-conversion) and the pulse duration is measured to be $\cong 170$ fs by autocorrelation (FWHM assuming Gaussian pulses). The pulse energy is controlled through the combination of a half-wave ($\lambda/2$) plate and a polarizing beam splitter (PBS). Then, the beam is injected into a stretcher arrangement consisting of two aluminum Littrow gratings (GR50-0616, Thorlabs) having 600 groove lines/mm to introduce negative group delay dispersion that results in chirping the laser pulses. The incident angle is 10° away from the Littrow angle. Then the stretched picosecond pulses can be sent to a homemade long-scan auto-correlator setup so that the pulse duration can be monitored at any time during the experiments. The range of accessible pulse durations is given in Fig. 1(b) where we show a calibration measurement as function of the parallel grating distance. The latter is continuously changed by placing the second grating (G2) and retroreflector on a motorized linear stage. The measured results are corresponding to a theoretical prediction based on the dispersion of a pulse with 35-nm reference bandwidth with the group delay dispersion (GDD) introduced. While it is not rigorous in our case, the incoming pulses are assumed to be transform-limited for simplicity in the calculations. As the distance between the two gratings (G1, G2) is varied from 25 to 145 mm, the GDD is varied from -1.4×10^5 fs² to -8.1×10^5 fs². According to this calibration and the scanned grating distance, any pulse duration from 4 ps to 21 ps can be applied for investigations in our Si irradiation experiments. This is done by inserting a mirror in the beam path to direct the beam into the interaction study setup. Experiments at pulse duration of 170 fs are also possible by simply bypassing the stretcher. For comparisons with long pulse regimes, we also couple an Erbium doped fiber laser to the setup (MWTech, PF1550) (not shown in Fig. 1(a)). The latter delivers pulses of 5 ns duration (FWHM) at the same wavelength of 1550 nm. A telescope

is installed on the beam path so that the beam diameter is identical to the femtosecond laser beam. For the material modification studies, the beam is focused using a microscope objective lens having a numerical aperture (NA) of 0.85 (LCPLN-IR 100× Olympus) in a 1-mm thick silicon wafer (intrinsic, <100> crystal orientation) mounted on a precise 3D actuator. The pulses are focused 300 μm beneath the surface to tentatively induce and detect in-volume damage. Spherical aberrations due to Air-Si mismatch are corrected with the aid of the adjustable correction collar of the objective. Fresh material zones are systematically irradiated during 1 second at 1-kHz repetition rate so that we compare the effects induced with 1000 pulses. We make this choice because of the sensitivity and resolution limitations of the infrared imaging (described below) system that we used for detection of potential modifications. While single shot modifications are hardly detectable, we found that 1000 shots are usually appropriate for the growth of the modifications and unambiguous detection. Applying the pulse sequences at 1 kHz repetition rate, we do not introduce any thermal accumulation effect to account in these experiments [5,23] and we only benefit from incubation in comparison from a single-shot study. For the investigations at different pulse durations, the maximum pulse energy impinging the Si target is 1.95 μJ (measured after the objective lens) corresponding to the maximum fluence of $\approx 86 \text{ J/cm}^2$ with theoretical beam diameter $1.22\lambda/\text{NA} \approx 2.4 \mu\text{m}$ in air. After irradiation, the samples are characterized by a home-built infrared microscopy imaging system with spatial resolution of $\approx 0.75 \mu\text{m}$ according to the magnification of the $\times 20$ objective lens (LCPLN20×IR, Olympus) and the pixel size of the InGaAs array (Raptor, OWLSWIR 640) used for the imaging arrangement.

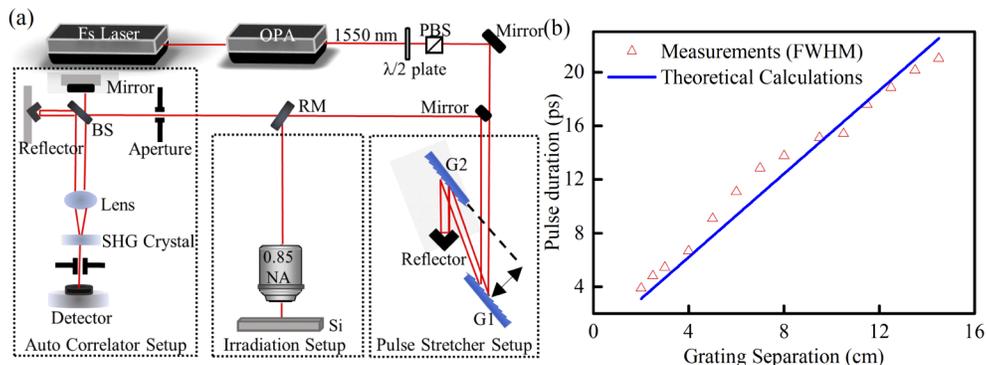


Fig. 1. (a) Schematic layout of the experimental setup for the study of the pulse duration dependence of laser-induced modification inside silicon. A femtosecond laser pumping an optical parameter amplifier (OPA) is used to deliver pulses of 170-fs duration at 1550 nm. The beam is injected into a grating pair to stretch the pulses. The distance between the two gratings can be continuously adjusted giving access to a range of pulse duration from 4 ps to 21 ps. The stretched pulses are focused by a microscope objective at 300 μm depth inside a Silicon sample. In addition, a home build long-scan auto-correlator is available with the aid of a removable mirror (RM) to characterize the pulses used for irradiation. (b) Serving as calibration, the measured pulse durations as a function of grating separation distance (marked by triangular symbols) are compared to a theoretical prediction accounting for the introduced group delay dispersion (marked by solid line).

For the characterization of the delivered fluence inside the samples (setup not shown), we have used the same Z-scan methodology as in our previous works [2,22]. A high-resolution imaging system equipped with an InGaAs camera is used to measure the transmitted beam at the back surface of a Si sample of given thickness. By moving the focused beam along the optical axis, we reconstruct the 3D fluence profile inside silicon as if the beam were focused at depth corresponding to the thickness of the sample used for the measurement. Because

such measurements are limited by the resolution of our measurement system built with a long working distance objective of 0.7 NA, we made these investigations for a beam focused with an objective lens (LCPLN-IR 20× Olympus) of NA=0.45. This objective is also equipped with a correction collar to deal beam free from spherical aberration in all cases. The modest NA in these experiments ensures that the fluence images are not low-pass filtered even in case of plasma defocusing. Unfortunately, such quantitative analyses are not accessible on the beam focused at NA=0.85 which is required for achieving modification. However, fluence clamping is found as a common feature in all ultrafast interaction inside silicon [2]. Then, the investigation of the pulse-duration dependence of the achievable fluence at NA=0.45 remains instructive on the general change of the conditions applied to matter when changing this parameter.

3. Results and discussion

3.1. Energy thresholds for bulk modification

First, we measured the damage energy thresholds (energy per pulse with 1000 applied shots) for bulk modifications at different pulse durations. An initial observation is that the different pulse durations are not equally appropriate to induce a modification inside Si. With the 170-fs pulse irradiation case, we cannot obtain any permanent change inside Si for all tested pulse energies, according to the detection by infrared microscopy. This holds when focusing with our higher magnification objective at NA=0.85 and obviously for lower focusing conditions (tested at 0.45 and 0.65 NA). In Fig. 2 we summarize the energy ranges for modification as a function of pulse duration. The absence of any damage at short pulse duration is symbolized by the ‘A’ region covering even shorter pulse durations by combining this observation with previous reports [2,9,21]. With the range of pulse durations accessible with the designed stretcher, we expect modification comes into picture as observed in some other previous works [5,23]. Interestingly, our shortest accessible pulse duration of $\cong 4$ ps (due to the minimum distance that can be set between gratings) leads to the same conclusion as for the femtosecond pulse irradiation. No bulk damage can be induced in the range of tested energy up to near 2 μ J. The incapacity or difficulty of achieving modification with short pulses have been also reported in different pulse duration regimes in previous works [5,21–23]. It is usually attributed to the clamping of delivered intensity inside Si due to detrimental nonlinear propagation effects increasing with pulse shortening [2,5,8]. The underlying mechanisms include progressive and strong two-photon absorption before the focus as well as Kerr effect and plasma screening or scattering effects preventing the pulse energy to be efficiently delivered near focus [2,8]. This yields that the non-damage region ‘A’ introduced above extends from the shortest possible pulses to pulse durations of more than 4 ps. When we progressively increase the pulse duration, we obtain bulk modification detectable with the infrared microscope. For pulses as short as 5.4 ps, we found conditions (at particular energy level) for which bulk modification can be systematically observed. Between 4 ps to 5.4 ps, it is also worth to note that sometimes it was possible to observe damage but it could not be associated to 100 % damage probability on a basis of irradiations repeated more than 10 times with identical conditions on different regions of the sample. This justifies the use of gradient in color in the representation of Fig. 2.

Another important aspect to consider here is that all attempts to achieve bulk modifications with NA=0.65 and below were unsuccessful. A consequence is that the description by a single energy threshold for modification depending on the other parameters (NA and pulse duration) is not appropriate as for usual material processing applications. There are thresholds to exceed on all these three parameters, at least. Here the NA threshold is in the range 0.65-0.85 and pulse duration threshold is near 5 ps. At pulse duration of 5.4 ps, we measure an energy threshold for modification at 84 ± 17 nJ as illustrated with the insert in Fig. 2 showing impacts at several pulse energies. Note that bulk modification energy thresholds (if so) are estimated by repeated irradiation of fresh regions of the sample with gradually increasing pulse energies

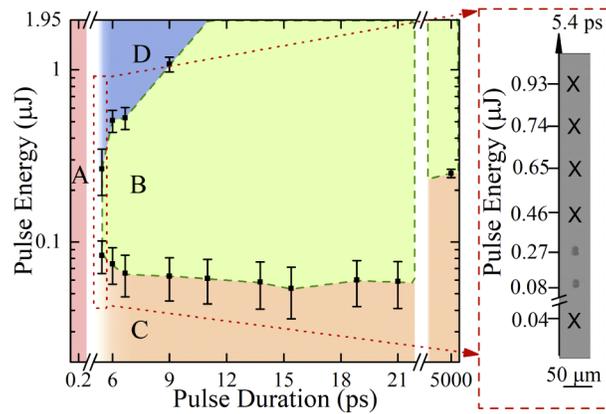


Fig. 2. Laser conditions for achieving micro-modification inside silicon with a 1550-nm wavelength laser focused with NA=0.85. The pulse energy and pulse duration ranges for modification with 100% probability are shown with region 'B' in green. The set of parameters leading to the absence of modifications are divided in 3 zones: 'A', the region where the pulse duration is too short to achieve modification independently of the pulse energy; 'C', a region below energy threshold for modification, and 'D', a region corresponding to high power regimes where the pulse energy is strongly delocalized setting leading to local energy densities in matter below conditions for modification. The inserted image (right part) shows the IR microscopy image of bulk modified spots systematically irradiated with 1000 pulses at different pulse energies (energy per pulse) for 5.4 ps duration pulses where non-damage parts are symbolized as cross marks.

until a modification is systematically observed. Then, the precision on the reported thresholds (shown by error bars in Fig. 2) is directly depending on the energy steps used in each damage test procedure. When we increase the energy continuously above this value, interestingly we find that at $0.27 (\pm 0.08) \mu\text{J}$, that the capacity to achieve modification vanishes. This means there is a relatively narrow energy range for which bulk modification can be inscribed as shown with the green region B in Fig. 2. This damage range can be directly visualized with the IR microscope image inserted in Fig. 2 (right part of Fig. 2) where we show repeated irradiation at different energy levels.

As the pulse duration increases up to 10 ps and more, we observe that such range of appropriate energy for bulk writing still exists, and it increases progressively. For pulse duration exceeding 10 ps, the threshold for delocalization (if any) shown with the transition between 'B' and 'D' regions becomes inaccessible for measurement due to energy limitations in the experiment. For technological consideration, the region 'B' of Fig. 2 is particularly interesting because it describes appropriate parameters for 3D writing defined by two measurable thresholds. The first threshold that we refer as 'low damage threshold' corresponds to the minimum incoming energy to deliver an energy density exceeding the modification the material. It corresponds to the boundary between 'C' and 'B'. The second 'high damage threshold', corresponding to the transition between 'B' and 'D', is the energy that should not be exceeded to avoid nonlinear delocalization inhibiting the pulse capability to inscribe damage in the material. Delocalization is attributed to a detrimental balance between nonlinear effects including Kerr effect and plasma defocusing. The latter can turn into re-localization at specific high-power levels as observed in a previous work [5]. In these new experiments, it is important to note that we have not observed re-localization. However, it is difficult to conclude from this comparison that whether the re-localization is general feature or not. Meanwhile, it is not observed here due to the limited

range of energy tested or it was something specific to previous work [5] where a stronger Kerr focusing contribution at 2- μm wavelength can dominate.

When looking at the evolution of these two thresholds, we see that the 'low damage threshold' first decreases from 0.084 to 0.065 (± 0.007) μJ when increasing the pulse duration from 5.4 to 6.7 ps and then keeps relatively stable. The increase of energy threshold with pulse duration shortening is consistent with nonlinear delocalization of the delivered energy that increases with pulse shortening and described in previous work [2,23]. However, it is very unusual to observe a pulse duration-independent threshold for modification in such a large range of durations in the picosecond regime (7 to 21 ps). By looking at the 'high damage threshold' evolution, we can safely confirm that nonlinear delocalization is rapidly decreasing for pulse durations in the picosecond range. Then, the nearly constant threshold suggests a direct compensation with the natural increase of required intensity for modification with pulse duration rise.

At this stage, it is important to highlight that the mapping of conditions for material processing shown in Fig. 2 strongly deviates to what is reported in surface [10] or bulk [12] machining studies in dielectrics where energy threshold for modification commonly increases with increasing pulse durations. Another difference is the the unusual region 'D' attributed to nonlinear delocalization in the dielectric studies.

Looking at the technological perspectives, these measurements confirm unambiguously that long pulse duration of few tens of picosecond are more favorable for 3D writing in Si. This is also confirmed by measuring the threshold with 5-ns pulse. The low damage threshold is found in this case to be around $0.25 \pm 0.02 \mu\text{J}$ under identical focusing conditions. While we should be careful with the comparison of two independent laser systems, this is relatively higher than what we measure in the picosecond range (see boundary between the region 'B' and 'C' in Fig. 2). This tends to show that when the duration of the pulse is long enough, the interaction becomes free from nonlinear propagation aspect due to reduced beam peak power and we recover a more usual pulse-duration scaling law for the threshold. Indeed, an increase with pulse duration following a square-root dependence is expected as soon as thermal dissipation is the limiting factor on the achieved energy density in materials like dielectrics and semiconductors [10,11].

3.2. Repeatability and reproducibility of induced modifications

In addition to the modification thresholds, we also found that the pulse duration can affect the reproducibility of induced modifications. In Fig. 3(a) repeated modifications produced to form a matrix inside silicon are shown by infrared transmission microscopy. On the X-axis of the matrix, the same irradiation conditions are repeated at a fixed pulse duration. As indicated with the label of the Y-axis, the pulse duration is changed between two consecutive lines. For direct comparisons, all the modifications are produced at a fixed pulse energy of 0.22 μJ , that is significantly above the low damage threshold for all pulse durations. According to the modest dependence of the energy threshold E_{th} (when accessible), this corresponds to an energy of about $\cong 3.5E_{th}$ for all pulse durations exceeding 6 ps leading to a directly relevant comparison in this range. For the shortest pulses, this energy is $> 2.5E_{th}$ that is also significantly above threshold conditions. From the Fig. 3(a), we can see that 100% damage probability is obtained for all pulse durations. This is a major difference with a previous work reporting only stochastic responses for all cases in the same range of pulse durations but using a lower NA (0.45) for focusing and a different laser technology [23]. In our case, we have checked that the change from NA=0.45 to NA=0.85 allows us to move from 0% probability to an existing set of parameters to achieve 100% probability. This likely indicates the important differences in the experimental conditions between the two studies. Interestingly, it shows also from our experiments the possibility of using tightly focused pulses from an OPA to reach local conditions in the matter significantly exceeding those for permanent modifications.

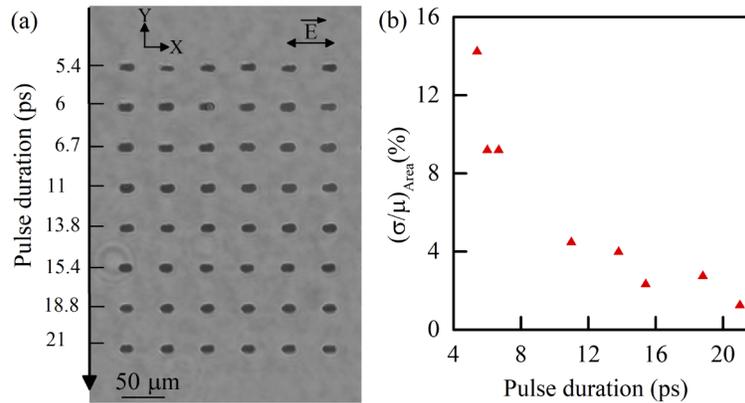


Fig. 3. (a) Infrared transmission microscopy image of micro-modifications produced with pulses tightly focused $300\ \mu\text{m}$ below the surface of a silicon wafer. As shown with the E vector orientation, the polarization is linear parallel to X axis. The modifications form an array where each line corresponds to repeated identical irradiations separated by $50\ \mu\text{m}$ at a given pulse duration indicated by the vertical axis. The pulse energy is $0.22\ \mu\text{J}$, significantly above the modification threshold energy for all tested pulse durations. (b) Stability of the apparent areas of the imaged modifications factor (zero standing for perfectly reproducible features) as a function of duration of the laser pulses used for writing.

Despite this repeatability on the occurrence of damage, we found differences in the reproducibility of the produced features. For the longest pulses, the apparent shape and size is nearly constant, but it is obviously changing when we compare the irradiated sites with the shortest pulses of 5.4-ps duration. To objectively confirm this observation, we quantify the reproducibility of produced features by introducing a stability factor being the ratio between the standard deviation σ of apparent modification areas in the image to the average area μ over the 6 modified spots for each pulse duration. The result of this analysis is shown in Fig. 3(b). We observe that at 5.4 ps the stability factor is modest near 14 % suggesting some instabilities in the energy delivery associated to strong nonlinear propagation effects. The latter become so high at high energy that they completely delocalize the light and prevent the occurrence of modification above the 'high damage threshold' reported in Fig. 2. Figure 3(b) also shows that the factor progressively decreases when the pulse duration increases and reaches 1.2 % for 21 ps. This must correspond to extremely reproducible local conditions in the matter. This is consistent with the idea that nonlinear propagation makes the situation more prone to instabilities. Indeed, the drop of the incoming peak power of a factor 4 with increased pulse duration in this experiment reduces these nonlinear effects which are detrimental for high energy density delivery. Another interesting observation not deeply studied here but clearly visible with Fig. 3(a) is a change from polarization orientation-dependent features with the shortest pulses to symmetric modifications produced with the longest pulses. Interesting these asymmetric structures are also reported in previous literature [1,25]. However, the underlying physical mechanisms remains an intriguing aspect which will need further investigations.

3.3. Pulse-duration dependence of laser energy densities in bulk Si

We can conclude from previous sections that pulses of few tens of picosecond are highly favorable for ultrafast laser 3D writing inside Si because it opens the possibility to produce extremely reproducible features and the energy threshold for modification is likely at its minimum. This optimum is obviously related to the delivered laser energy density. Also, as we did in previous works concentrating on the focusing NA [2], we have measured the 3D fluence distributions

inside Si for different pulse durations. The details of the experimental procedure to retrieve the distributions from a Z-scan measurement are available in Ref. [2]. The Fig. 4(a) shows cross-sections along the optical axis (propagation from top to bottom) of the fluence distributions inside Si near focus ($z=0$) for two different pulse durations at three different incident pulse energies on the sample. The calibration to represent the delivered fluence inside Si accounts for Fresnel's reflection at Air-Si interface. Due to imaging limitations, we investigate here the delivered fluence for our beam focused with $NA=0.45$. This focusing condition is inherently below permanent modification threshold. Thus, there is no structural modification of the material affecting the beam propagation in these images acquired by accumulation on an InGaAs array over many pulses ($> 10\,000$ at 1 kHz).

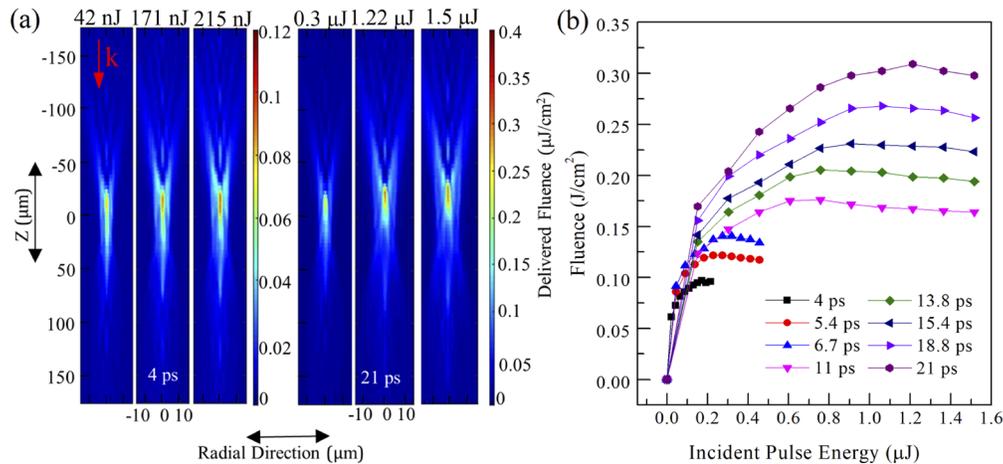


Fig. 4. Delivered fluences in Si for a beam focused with $NA=0.45$ (a) Cross-sections of 3D spatial fluence distributions in Si acquired by Z-scan imaging at three incident pulse energy conditions for 4-ps and 21-ps pulses. The propagation direction is indicated by the wave vector (k) and the geometrical focus corresponds to $z=0$ (b) Peak delivered fluence found in the distributions as a function of incident pulse energy for pulses with duration gradually varied from 4 ps to 21 ps.

First, we observe from the comparison between 4 ps and 21 ps at different incident pulse energy levels that the fluence profiles exhibit diffraction patterns [2] due to overfilling of the entrance pupil of the objective and all are relatively similar. We simply observe a sharp tail in the back focal region more pronounced for the higher peak power reached with 4-ps pulses. This leads to a feature somehow similar to what is found for a beam focused with spherical aberrations [27] which may be still present throughout the measurement despite an implemented compensation. The beam distortions of the interacting beam however remain not so severe because the images show that the focusing power of the beam is preserved. The overall shape of the focused beam distribution and its position with respect to the geometrical focus is kept similar within around 15 μm range for the different incoming laser conditions.

When looking at the fluence scales of the distributions, it is worth noting that the delivered peak fluence is not increasing proportionally with the incident pulse energy. To access more details, we plot the delivered peak fluence as a function of the incident pulse energy delivered to the sample in Fig. 4(b). We can see a clear saturation of the delivered pulse fluence with 4-ps pulses. The peak fluence increases first linearly with respect to incident pulse energy, then it saturates at a maximum fluence value of $\cong 0.1$ J/cm² when the delivered pulse energy to the sample reaches about 0.17 μJ. We have taken this energy as a reference for saturation (E_{sat}) for comparison with the response at other pulse durations exhibiting also saturations as shown in Fig. 4(b). The

results are very similar to our previous results when measuring the delivered fluence for shorter pulses down to sub-100-fs pulses [2–5]. Supported by simulation results, the saturation was mainly explained by energy delocalization attributed to nonlinear effects, including two-photon absorption and plasma diffraction and screening effects in the pre-focal region. Despite similar saturation, curves are observed for pulses longer than 4 ps, we can note that the saturation level increases evidencing higher energy delivery. From 4 ps to 21 ps, the incident pulse energy leading to fluence saturation increases from 0.17 μJ to 1.22 μJ . As the nonlinear effects are associated to a critical intensity and/or peak power, they are logically triggered at higher energies when increasing the pulse duration. However, the detailed characteristics of the fluence distribution do not rigorously indicate the same nonlinear propagation signatures. Looking back to the 3D fluence profiles shown in Fig. 4(a), the three different energies are selected for each pulse duration so that they correspond to nearly identical ratios with respect to the saturation energy E_{sat} . The pulse energies correspond respectively to 0.25, 1 and 1.25 E_{sat} . Also, the differences pointed out before, even if it is modest, indicates that the origin of the saturation does not strictly follow an identical change of propagation at a given peak intensity/power.

Another interesting observation with Fig. 4(b) is that the curves obtained for different pulse durations do not exhibit only a strict saturation, they tend to show slightly decreasing fluences at high energy levels. The magnitude of the drop may be different for the NA=0.85. However, combined to potential effects affecting the pulse in the temporal domains [28], it must represent an important aspect to explain the 'high damage threshold' found in the damage study (see section 3.1). This defined an upper limit of the modification energy range (when existing) because higher energy above saturation degrades the energy deposition conditions in bulk material precluding locally the delivery of a sufficient energy density to modify it permanently [5].

Despite the systematic saturation behavior, it is obvious from Fig. 4(b) that the delivered peak fluence increases with pulse duration. To visualize this trend, we show in Fig. 5 the maximum delivered peak fluence (taken at E_{sat}) as a function of pulse duration. To serve as guideline for future development, it is interestingly to note the nearly linear dependence to this parameter in the range of tested pulse durations. This information adds to our previous report on a linear dependence of this observable to the NA [2]. It is important because, it indicates that it becomes possible to extrapolate the maximum fluence accessible for any pulse duration and NA combination from a single measurement. However, it remains hard from this knowledge of the delivered fluence to be fully aware of how far the conditions to the permanent modification are. This is because not only the delivered fluence is changing with pulse duration but also the fluence threshold for modification.

To address this question, we have measured the laser induced damage threshold (LIDT) on the sample surface using a standard single-shot methodology [29] with the same laser and we show on Fig. 5 (right axis) the ratio of delivered fluence measured in the bulk to the surface LIDT. This shows a ratio of about 0.1 and nearly constant in the range of pulse durations accessible for this experiment. As bulk modification thresholds are usually higher than surface thresholds [30,31], this indicates conditions far from modification thresholds. In addition, Fig. 5 shows that the conditions in the bulk are not significantly approaching the requirements for permanent modification when increasing the pulse duration in this range. It remains that we cannot envision a damage threshold fully independent on pulse duration and this observation is likely a specificity of the range of pulse duration tested here. Indeed, when the pulse duration largely exceeds the lattice thermalization time (through electron-phonon coupling) on the order of a few picoseconds, thermal runaway assists energy deposition by bandgap closure (thus facilitating the reaching of energy threshold modification of the material in the long pulse regime). Ultimately, the energy deposition volume must be governed by thermal diffusion implying that the measured LIDT become proportional to the square root of the pulse duration. Assuming the linear increase of deposited fluence still applies for pulses longer than those in the tested range, this means that the

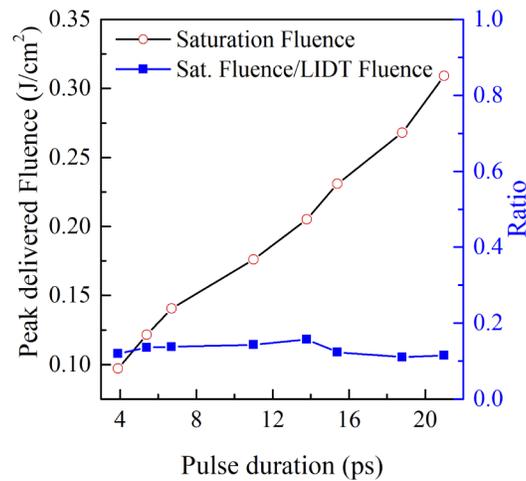


Fig. 5. Maximum achievable fluence (left axis) in Si with NA=0.45 and ratio to the permanent damage threshold (right axis) as a function of pulse duration.

ratio shown in Fig. 5 must inevitably increase for longer pulses and reach unity level when the delivered fluence and LIDT curves cross each other. This picture is consistent with our previous studies and others [15,17,19] reporting modifications in Si with NA=0.45 for a pulse duration of about 5 ns. This proves that the ratio exceeds 1 in the nanosecond regime and a ratio reaching the unity level for minimum pulse duration between 20 ps and 5 ns.

4. Conclusion

In conclusion, we have investigated the pulse duration dependence of the conditions applied in Si with tightly focused pulses. A major conclusion is the achievability of 3D laser writing at NA=0.85 with pulses of 5.4 ps and more as shown with Fig. 6 summarizing the results of this study. With shorter and more intense pulses, the detrimental nonlinear propagation effects become too strong to exceed the modification threshold, independently of the pulse energy. While we concentrated here on Si, we have tested the response of other semiconductor materials (including ZnS, ZnSe, GaAs). Future works should be based on the detailed response on these materials, but we observed that this 'Abnormal' response is not a specificity of Si but can be likely generalized to bulk interactions in narrow gap materials. Obviously, we are not the first to achieve bulk writing in Si, but it is striking to note that the data in the intermediate picosecond regime are scarce and somehow contradictory [5,23]. Over the last decades, important bulk Si modification technologies such as stealth dicing, laser assisted etching and some other photonics applications have been developed with nanosecond pulses that allow to readily induce internal damage in a Si wafer [15–17,19,32]. Also, it is only in the last few years that the possibility to achieve refractive index engineering in Si has been demonstrated with sub-picosecond pulses [2–4]. This is because important efforts have been needed to circumvent the detrimental nonlinearities observed also in this work with the shortest applied pulse. One solution which has been identified is the use of extreme NA values [2]. It is interesting to note that in the same way we found in a previous work a NA value of near 3 that must be exceeded to achieve modification with sub-picosecond pulses, we find here that a pulse duration threshold of about 5 ps must be exceeded to achieve modification at NA=0.85.

This leads to a situation where the occurrence of modification cannot be simply described by a pulse energy threshold depending on the other parameters. The NA and pulse duration are two other parameters on which some threshold responses also apply. Taken together, the

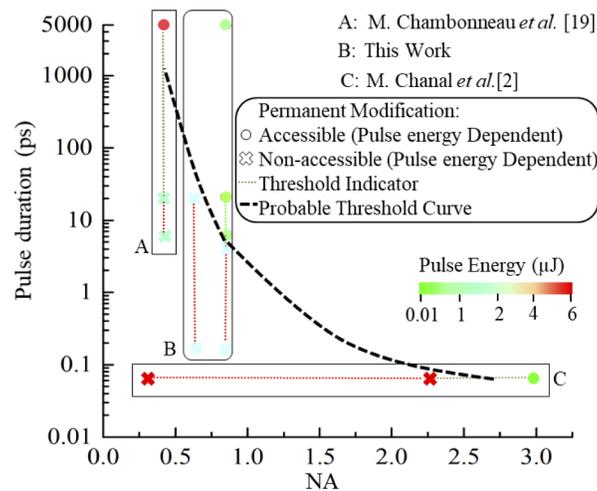


Fig. 6. Thresholds in pulse duration and NA that must be exceeded (in addition to pulse energy threshold function of the situation) to achieve permanent modification in bulk Si with focused laser pulses at wavelength in the range 1.3-1.55 μm . We report successful and unsuccessful attempts of this study by respectively hollow circle and cross symbols filled into respective pulse energy color scale value. The successful attempts obtained with more modest NA in the nanosecond regime (shown inside square box A) [19] and extremely high NA for the shortest pulses (illustrated under square box C) [2] are complemented by the current work (implemented under box B). In this way, we show with the dashed black line the probable threshold conditions which must be exceeded to achieve modification.

threshold conditions are defined by at least a triplet of parameters defined by the incoming pulse energy, NA and pulse duration. In that respect, our work makes a crucial step in mapping the required conditions for internal silicon structuring. To illustrate this advance from this study, we summarize in Fig. 6 by putting hollow circle and cross symbols (solid circles) and unsuccessful attempts (crosses) with different the NAs and pulse durations tested. Thanks to the ability to continuously tune the pulse duration, we have positioned very precisely a NA and pulse duration combination threshold in the picosecond regime. By reporting also on the same graph the extreme NA requirements with femtosecond pulses [2] and some successful conditions with relatively modest NA in the nanosecond regime [19], we can also show the typical dependence to these parameters of modification threshold by a dashed black line on this graph. This must represent an important guideline for the developments of ultrafast laser 3D writing technologies which remains challenging but holds great promises to the future electronic industry.

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Disclosures

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