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Predictable surface ablation of dielectrics with few-cycle laser pulse even beyond air ionization

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We study surface ablation of dielectrics with single-shot few-cycle optical pulse (≈10 fs) in air, at intensities below and above the onset of air ionization. We perform 3D analysis and careful calibration of the fluence distribution at the laser focus, spanning from linear– to nonlinear– focusing regimes, enabling to thoroughly characterize the severe limitation of the fluence delivered onto the sample surface upon increase of incident pulse energy. Despite significant beam reshaping taking place at high fluence, we demonstrate that it is nevertheless possible to confidently predict the resulting crater profiles on fused silica surface, even in the regime of filamentation. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4960152]
At low incident energy, we measure Gaussian beam distribution and propagation [Fig. 1(a)]. The position $z = 0$ on the optical axis corresponds to the focal plane, referred to as “linear” focus, in the absence of nonlinear propagation in air. The fluence is defined by $F_G(r,z = 0) = F_0 \exp[-2r^2/w_0^2]$ with $F_0 = 2E/\pi w_0^2$ the peak fluence ($E$ is the pulse energy) and $w_0 = 11.0 \mu m$ the measured beam radius at $1/e^2$. The sample is then precisely located in the focal plane using combined energy-scan and z-scan procedures with an accuracy of $50 \mu m$, much better than the Rayleigh range $z_R = \pi w_0^2/ M^2 \lambda = 396 \mu m$ (with measured $M^2 = 1.2$).

For Gaussian beam, and assuming deterministic characteristics of interaction, the diameter $D$ of the crater should satisfy the relationship\(^{(13)}\) (expressed either in terms of fluence or energy)

$$D^2 = 2w_0^2 \ln(F_0/F_{th}) = 2w_0^2 \ln(E/E_{th}).$$

(1)

The ablation threshold $F_{th}$ is defined by $D^2 = 0$. Figure 2 shows that the experimental data are fully in accordance with this equation only for low incident energies, until $E_{NL} = 7.5 \mu J$ (corresponding intensity $I_{NL}^2 = 1.65 \times 10^{14} \text{Wcm}^{-2}$). As expected, this value is slightly above the intensity we previously determined,\(^{(12)}\) the small difference being related to the observable used for defining it. In our former experiment, we indeed measured the energy above which the beam focal plane begins to move out of the linear focus while here we observe the consequences of such displacement on an outcome of the interaction, e.g., the ablation diameter.

Importantly, this enables to retrieve the ablation threshold of the fused silica sample ($F_{th} = 1.8 \text{~Jcm}^{-2}$, expressed in terms of peak fluence), which is well below the energy level corresponding to the onset of nonlinear effects. In addition, the slope of the fit gives the beam waist, $w_0 = 11.3 \mu m$, which is in excellent agreement with the measured value. Thus, up to $E = E_{NL}^3$ (corresponding to $F = 4.1 \text{~Jcm}^{-2} = 2.3F_{th}$), the crater diameter is completely and easily predictable. This is a finding of high practical importance: indeed, working under vacuum environment is not absolutely mandatory when using $\sim 10$ fs pulses, provided that the fluence is properly adjusted. A range of fluence close to threshold exists such that ablation in air is not hampered.

For incident energies above $E_{NL}^3$, crater diameters deviate from Eq. (1), with dimensions larger than expected. Beam analysis [Figs. 1(b) and 1(c)] indeed reveals how the nonlinear propagation in air changes the beam properties prior the linear focus. Note that for high energies the accuracy of the imaging system is somewhat lowered in the region up to the nonlinear focus only, because in this case the beam is measured through a medium (air) developing optical aberrations. However, in our experiment, the main interest is to know the details of the beam in the plane of the sample, thus in an observation zone where the imaging set-up is free of any optical aberrations. At this point, let us specify also that our aim is not to perform a dedicated study of beam propagation in air under external focusing, and not to characterize the density of the plasma generated in air but rather to link ablation results to beam analysis. In particular, self-focusing and air ionization cause changes in the $z$-position of the actual focus, as well as on further propagation properties. As a result, the actual beam at the sample surface (still located in the linear focus plane) is severely distorted, so that ablation may result in craters of completely different characteristics. The crucial point for prediction of crater diameter is the ability to accurately depict the

FIG. 1. 2D mapping of the reconstructed fluence along beam propagation for three different energies corresponding to the three regimes of propagation identified (see after). The fluence is calibrated by taking into account the energy conservation and the measured beam size, considering Gaussian beam propagation for cases (a) and (b) and superposition of Gaussian and superGaussian beams for case (c). The position $z = 0$ (“linear” focus) is indicated by the dashed line.

FIG. 2. Measured ablated diameters squared $D^2$ as a function of incident energy (logarithmic scale). The numerical fit (continuous line) is obtained using the equation $D^2 = 2w_0^2 \ln(E/E_{th})$ for data corresponding to $E < E_{NL}^3$. The ablation threshold $E_{th} = 3.6 \mu J$ is obtained when the fit reaches $D^2 = 0$. The data of ablated diameters are averaged over 10 points, using confocal microscopy. Three craters and 1-D profiles are shown in the three different regimes.
In fact, such beam profile reproduces very well theoretical results from propagation in filamentary regime under tight external focusing. Indeed, Chin et al.\textsuperscript{14,15} have shown that in filamentary regime the intense laser beam is composed of single fundamental mode, the filament core, and all other higher order modes that form the background reservoir.\textsuperscript{16} The role and details of the energy reservoir were shown and clarified in Refs. 17–19. Following Ref. 17, the energy reservoir is expected to be (i) much larger than the core and (ii) to contain the major part of the pulse energy. Thus, the appearance of such beam distortions upon increase of incident energy highlights the need to re-define properly the fluence. With the aim of providing straightforward assessments and predictability of crater dimensions for ablation experiments, we now demonstrate that such fluence profiles can be well fitted by a combination of superGaussian functions (SG) of order \( n \)

\[
F(r) = \sum_{n} E_{x_n} \left( \frac{2^{1/n} \Gamma(1/n)}{\pi^{1/n} n} \right) \exp \left[ -2 \left( \frac{r}{w_n} \right)^{2n} \right]. \tag{2}
\]

The normalization factor (in bracket) depends on the order \( n \) and \( \Gamma(x) \) is the Euler Gamma function. For every beam that we measured for a given energy \( E \), the free parameters of the numerical fit are: the order \( n \) of each participating SG function (note that \( n = 1 \) corresponds to a Gaussian function), the radius \( w_n \) at \( 1/e^2 \), and the weighting coefficient \( E_{x_n} \) ensuring that the 2D integration over \( r \) of the fluence function \( F(r) \) equals the energy carried by the beam. Note that we neglect the small loss of energy due to plasma creation in air, since we measured in previous work\textsuperscript{12} that it is limited to \( \sim 1\% \), in agreement with recent numerical predictions\textsuperscript{20,21} in such sharp focusing conditions. In fact, the numerical fitting of the beam enables to evaluate the peak fluence value (used to calibrate Fig. 1), which is an essential data to know for predicting the crater diameter.

For illustrative purposes, we focus now on the case \( E = 30.2 \mu J \). Figure 3 shows an excellent agreement between the experimental beam profile on the sample and the numerical fit obtained from Eq. (2), corresponding to the superposition of a Gaussian function (\( n = 1 \)) and two SG functions (\( n = 3 \) and \( n = 4 \)). The retrieved fitting parameters fully agree with Ref. 17: we obtain \( w_1 = 14 \mu m \), \( w_3 = 31 \mu m \), and \( w_4 = 33 \mu m \); and \( x_1 = 0.35 \), \( x_2 = 0.60 \), and \( x_4 = 0.05 \), thus providing quantitative measurements of the respective dimensions and ratios of energy contained in the reservoir and in the main peak.

In Fig. 3, \( w_3 \) and \( w_4 \) are the Gaussian and superGaussian radii at the peak fluence, respectively. The retrieved parameters are: \( w_1 = 14 \mu m \), \( w_3 = 31 \mu m \), \( w_4 = 33 \mu m \), \( x_1 = 0.35 \), \( x_2 = 0.60 \), and \( x_4 = 0.05 \), thus providing quantitative measurements of the respective dimensions and ratios of energy contained in the reservoir and in the main peak.
Finally, and thanks to the normalization factor in Eq. (2) issued from the fits for every case of energy investigated, the peak fluence gets calibrated and therefore so does the local fluence function $F(r)$ for every location $z$ along propagation. Two important values are the peak fluence $F_{0}^{\text{NL}}$ attained at the nonlinear focus during propagation, and the real peak fluence at the sample surface $F_{0}^{\text{sample}}$. These values are also reported in Table I for three energy cases illustrating the three regimes identified. Now, considering our whole set of experimental data, Figure 4 plots the real peak fluence at the sample surface $F_{0}^{\text{sample}}$ upon increase of incident energy. It reveals the appearance of a limitation of the fluence “seen” by the sample surface, starting from the energy $E_{\text{sat}}$, and followed by a complete saturation for $E > E_{\text{ioniz}}$ (around $F_{0}^{\text{sample}} \sim 51 / \text{cm}^2$ in our experimental conditions). Therefore, for incident energy above $E_{\text{ioniz}}$, the real fluence delivered on the target is strongly reduced compared to the fluence $F_{0} = 2E/\pi w_0^2$ calculated without nonlinear effects in air (i.e., considering that the beam is still Gaussian-shaped with $w_0$ size), therefore markedly modifying the ablation result that could be expected. We want to stress this particular point, since omitting to take it into account would yield strong misleading information when interpreting ablation experiments with few-cycle pulses in air.

Nevertheless, even if the beam is distorted (high energy case), the crater quality is maintained as shown by confocal microscopy (see 1D-profiles in Fig. 2). Surprisingly, high quality ablation with strongly distorted beams is therefore still possible. Moreover, thanks to the accurate beam characterization and proper fluence calibration, the crater diameter can even be predicted. Indeed, as we know the threshold peak fluence $F_{th}$, it is straightforward to extract the beam diameter at local fluence equal to $F_{th}$ (see Figure 3). As illustrated in Table I, the matching with measured crater diameters is excellent.

We also point out that at high energy, the beam undergoes spectral and temporal distortions, as it was measured in Ref. 12, leading, respectively, to enlargement and blue shift of the spectrum (due to air ionization) and a slight reduction of the pulse duration taking place in the central zone of the beam. However, spectral and temporal reshaping of the beam does not impede the prediction of the ablated crater based on the calibration of the fluence as well as its regularity and overall quality as shown in Fig. 2.

In conclusion, we have investigated if working in air may hamper surface ablation of dielectric materials by a tightly focused few-cycle pulse. We analyzed the transition from linear–to nonlinear–focusing regimes, including air ionization, therefore identifying three working ranges limited by $F_{th}$ and $E_{\text{ioniz}}$. The first important conclusion is that a fluence range exists ($F_{th} < F < 2.3F_{th}$) for which propagation is free of any nonlinear effects, thus permitting to benefit from the advantages of few-cycle pulses for ablation. Second, upon increase of energy, we demonstrated by accurate characterization and proper calibration of 3D fluence distribution that the actual fluence delivered on the target surface is strongly limited, due to severe reshaping of the beam (including filamentation). Nevertheless, high-quality and fully predictable ablation is still reached. This work therefore provides useful information to the laser user (including also the possibility to translate the sample at the nonlinear focus location) to extend the capabilities of few-cycle pulses in a large range of operating fluence, far beyond the natural limit imposed by air ambiance.

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References


