D-Scan Measurement of the Ablation Threshold and Incubation Parameter of Optical Materials in the Ultrafast Regime

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Abstract: We extend a simple method to measure the ultrashort ablation threshold of solid samples, the D-Scan, to take into account the pulses superposition. Its simplicity allows many measurements to be quickly performed under various superposition conditions, providing the sample incubation parameter. Preliminary results obtained for dielectrics indicate that the ablation threshold and incubation parameter increase with the bandgap energy.

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Introduction

The interaction of femtosecond pulses with matter occurs mainly through the coupling of the electromagnetic field with the lattice electrons, conferring to the ultrashort pulses ablation a nonselective character [1]. Due to this interaction, the only parameter needed to be known when machining a material with ultrashort pulses is the material ablation threshold, $F_{th}$. On dielectrics and semiconductors, the ultrashort pulses ablation is a nonlinear process initiated by electrons being freed from the matrix by multiphoton ionization [2] or by tunneling induced by the laser field [3], which evolves into a buildup of free electrons and culminates in the ablation when the electron density reaches a critical value [4]. In a given material, the ablation threshold can depend on the presence of impurities, defects, excitons, etc. [5], which either create intermediate levels in the bandgap or modify the local electronic density. As a consequence, electrons are freed more easily than in the ideal material, and the ablation happens at lower $F_{th}$ values. The defects can be intrinsic to the material, or externally originated, such as laser-created color centers [6], and in this case, when processing solids with superimposing pulses modifications can be induced in the material, lowering the $F_{th}$ for subsequent pulses. These cumulative phenomena fall under the classification of incubation effects [7, 8], and the ablation threshold fluence modifications induced by them must be taken into account when machining a material [9].

The measurement of the ablation threshold is usually done using the “zero damage” method [10], which requires a complete knowledge of the laser beam geometry, a precise and stable positioning of the sample under study, and measurements that can involve the use of an electronic or atomic force microscope to determine the ablation spot size. This technique can be experimentally demanding, and the determination of incubation effects requires its repetition for the superposition of many pulses, which can take a long time.

The Diagonal Scan (D-Scan) technique [11, 12], introduced by us, is a simple and quick method to measure the ultrashort pulses ablation threshold. In this method, the sample under study is diagonally moved across the waist of a focused laser beam, and the profile shown in Fig. 1 is etched in its surface.

It can be shown [11] that the ablation threshold is given by:

$$F_{th} = \frac{E_0}{e\rho_{\text{max}}^2} \approx 0.117 \frac{E_0}{\rho_{\text{max}}^2}$$  \hspace{1cm} (1)

where $E_0$ is the laser pulse energy and $\rho_{\text{max}}$ can be measured in the sample under an optical microscope.
To observe the incubation effects, the pulse superposition that etches the profile at \((\chi, \rho_{\text{max}})\) must be known. This superposition can be considered to be the ratio between the summation of the intensities produced at \((\chi, \rho)\) by every pulse that hits the sample during its movement across the beam waist, and the intensity generated by the pulse centered at \((\chi, 0)\). Under this assumption, it can be shown [13] that the superposition \(N\) produced at \((\chi, \rho_{\text{max}})\) is given by:

\[
N = \vartheta_3(0, e^{-\left(\frac{\chi}{\rho_{\text{max}}}\right)^2}),
\]

where \(\vartheta_3\) is the Jacobi elliptic theta function of the third kind [14], \(f\) is the laser repetition rate and \(v\) is the sample transversal translation speed [12].

**Results**

To validate expression (2) for the pulses superposition, a Ti:Sapphire CPA system (Femtopower Compact Pro HP/HR, from Femtolasers) was used to perform ablation threshold measurements using both the traditional “zero damage” and the D-Scan methods. Pulses centered at 785 nm, with 200 µJ maximum energy and 100 fs of duration (FWHM) on a 4 kHz maximum repetition rate pulse train were employed, focused by a 38 mm focal length lens, to determine the \(F_{\text{th}}\) dependence on the pulses superposition on BK7 samples [13]. The results are shown in Fig. 2 as red circles for the “zero damage” method and as black squares for the D-Scan using 3 different energies. The good results agreement validates the D-Scan method to measure accumulation [5, 7], leading to a constant value of parameters obtained from the fittings are shown in Table 1, together with the bandgap energy of each material.

The incubation effects in dielectrics can be described by a model that considers a saturation of the defects accumulation [5, 7], leading to a constant value of \(F_{\text{th}}\) for the superposition of many pulses:

\[
F_{\text{th}}(N) = F_{\text{th}}(\infty) + (F_{\text{th}}(\infty) - F_{\text{th}}(1))e^{-k(N-1)},
\]

where \(k\) is the sample incubation parameter, \(F_{\text{th}}(N)\) and \(F_{\text{th}}(1)\) are the ablation thresholds for \(N\) pulses and for a single pulse, respectively, and \(F_{\text{th}}(\infty)\) is the ablation threshold for infinite pulses, when saturation occurs. Figure 3 shows the ablation thresholds measured for three common optical materials, fused silica, sapphire and BK7, as a function of the pulses superpositions, and Eq. (3) fitted to each data set. For these measurements, the same Ti:Sapphire laser was used, but generating 25 fs pulses. For each sample, all the data were collected in less than 1 hour. The parameters obtained from the fittings are shown in Table 1, together with the bandgap energy of each material.
Table 1. Fit parameters for the data shown in Fig. 3 and bandgaps.

| Sample   | $F_{th,1}$ (J/cm$^2$) | $F_{th,\infty}$ (J/cm$^2$) | $k$      | bandgap (eV) |
|----------|------------------------|-----------------------------|----------|---------------|
| BK7      | 1.5(2)                 | 0.46(2)                     | 0.015(4) | 4.3 [15]      |
| Sapphire | 4.2(1)                 | 1.49(4)                     | 0.022(3) | 8.8 [16]      |
| Suprasil | 5.1(1)                 | 1.92(5)                     | 0.064(8) | 9.0 [16]      |

Fig. 4. Ablation thresholds (circles, left side scales) and incubation parameters (blue squares, right scale) dependence on the bandgap energy.

The results presented in Fig. 4 show that the ablation threshold, for single shots and also for the superposition of infinite pulses, increases with the bandgap growing. This behavior is expected once the creation of seed electrons demands that more energy is used to overcome larger bandgaps, increasing the total energy demanded for ablation. As for the incubation parameter, a larger value means a higher defect accumulation rate, and the observed increase can imply that the defect recombination rate is smaller in materials with bigger bandgaps due to the higher barrier to be overcome. Nevertheless, more data from dielectrics with bandgaps in the intermediate range (5-8 eV) is needed to strengthen these conclusions.

Conclusions

The D-Scan method, whose one of the strengths is its simplicity that allows many measurements to be quickly made, was validated for the measurement of the ablation threshold for the superposition of ultrashort pulses by comparing its results to the ones obtained by the traditional method. This method was then used to determine the incubation parameter for optical materials, indicating that those increase together with the material bandgap.

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