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Laser ablation of dielectrics with temporally shaped femtosecond pulses

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A significant improvement in the quality of ultrafast laser microstructuring of dielectrics is demonstrated by using temporally shaped pulse trains with subpicosecond separation. The sequential energy delivery induces a material softening during the initial steps of excitation changing the energy coupling for the subsequent steps. This leads to lower stress, cleaner structures, and provides a material-dependent optimization process. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1432747]

Femtosecond lasers have been recognized as potential tools for micromachining due to advantages compared to nanosecond processing,[1–3] in particular localized material removal and reduced heat-affected zones. Ultrafast lasers have also opened new opportunities in exploring the basic aspects of laser-induced damage and ablation.[4,5] Femtosecond irradiation induces a fast energy deposition into the electronic system, temporally decoupled from the relaxation to the lattice, simplifying fundamental analyses. Several studies[6,7] have indicated specific paths for energy transfer to the lattice depending on the strength of the electron–lattice interactions. These studies have shown fast trapping of free electrons in oxides with strong electron-phonon coupling (SiO_2) within 100 fs, while for other oxides like α-Al_2O_3 or MgO, the electrons remain quasifree for tens of picoseconds. Knowledge of the individual response times of the materials establishes a guideline for the use of temporally tailored pulses to optimize the structuring process with respect to the reduction of the residual damage. The emphasis of this letter is to illustrate the potential of temporally shaped laser pulses for microprocessing.

Studies of ablation mechanisms on dielectrics with subpicosecond, near-infrared laser pulses have shown several characteristics:

The laser energy results primarily in a localized electron-hole plasma formed by multiphoton seeded avalanche multiplication,[4,5] and further affected by photoemission, surface charging, electron drift, trapping, recombination, and diffusion.

Following incubation, the absorption changes from being localized in the electron plasma in the initial gentle ablation phase to absorption in an extended defect-affected region in the strong phase.[8]

An impulsive, electrostatic breakup of the surface in the gentle phase[8] leads to high kinetic energies and charge-scaled momenta for the emitted ions. Kinetic energy decreases in the strong phase where thermal effects prevail.[8]

There is a material-dependent time for releasing the electron energy to the lattice.

Temporally shaped femtosecond laser pulses would thus allow exploitation of the dynamic processes and control thermal effects to improve structuring.

Dielectric samples were irradiated with pulses from an 800 nm/1 kHz Ti:sapphire laser system delivering 90 fs pulses at 1.5 mJ. A novel technique for dynamic phase modulation was applied for quality-oriented microprocessing. A 2×128 pixel liquid crystal (LC) spatial modulator was inserted in the Fourier plane of a zero-dispersion stretcher. The use of LC modulators in spatially dispersed beams allows phase/amplitude modulation (by controlled retardation and attenuation at each pixel position in the frequency space) and, in turn, tailoring of pulses to desired shapes.[9] The modulator was inserted after the oscillator, the amplifier being seeded with the phase-modulated beam. The temporal profile of the modulated sequence was measured by second order cross correlation with a nonmodulated pulse deflected from the oscillator prior to phase modulation and seeded in the amplifier 800 ps behind.

Previous time-resolved experiments on ion emission from femtosecond laser irradiated sapphire (Al_2O_3) and fused silica (a-SiO_2)[8,10] indicated two regions of interest:

A fast, nonthermal feature of subpicosecond duration, originates in a Coulomb explosion of the surface.[8] This was seen only for dielectrics, for metals and semiconductors fast neutralization of the excess charge occurs before lattice breakup.[10]

A slower feature follows due to a thermal mechanism on a time scale given by the electron-phonon coupling and heat transport.[8] The thermal peak onsets at shorter times for SiO_2 compared to Al_2O_3 due to efficient electron self-trapping and increased electron-lattice coupling.[6]

These experiments illustrate the importance of the dynamics of the energy transfer to the lattice on material-dependent time scales. An informative experiment that renders access to electronic behavior is the study of laser induced optical damage with temporally tailored pulses. Figure 1(a) depicts the behavior of the damage threshold

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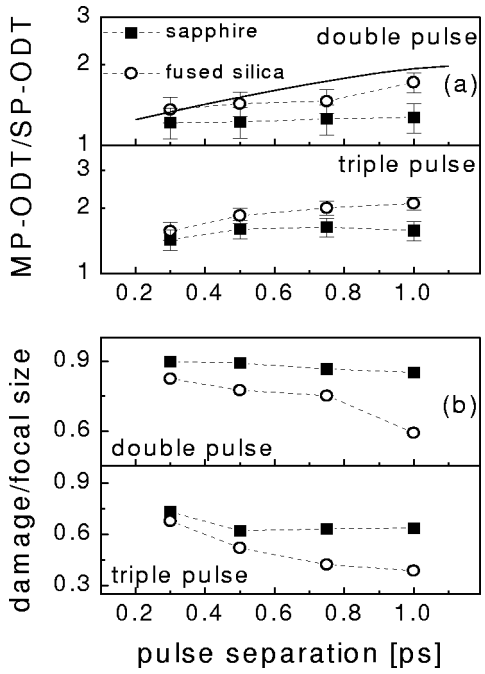

FIG. 1. (a) Optical damage thresholds for Al_2O_3 and a-SiO_2 for double and triple-pulse irradiation (MP-ODT, considering the total energy of the sequence) relative to the single pulse damage (SP-ODT) at different pulse separation times. The solid line is calculated for a-SiO_2 using Eq. (1). (b) Multipulse damage dimensions relative to the focal spot for Al_2O_3 and a-SiO_2 for different pulse separation times and fixed fluence. Smaller structures are obtained when pulse trains are used.

for single sequences of identical, double and triple pulses of different separation times, for Al_2O_3 and a-SiO_2, providing insights to the distinct trapping times.

The results (based on the zero-value extrapolation of the damage dimensions measured at different fluences) indicate a strong dependence of the multipulse optical damage threshold (MP-ODT) for a-SiO_2 on the pulse separation time [the multipulse threshold is given relative to the single pulse damage threshold (SP-ODT)]. The reported fluences include the contribution of all the pulses in one train. The increase in the damage threshold for a-SiO_2 is due to a loss in the electron population generated by the first pulse, affecting the subsequent ones. Constant behavior has been observed for Al_2O_3, where 100 ps electronic decay time[6] is too long for the time scale involved.

A similar dependence is observed in the multipulse damage dimensions [Fig. 1(b)]. A tendency towards smaller spot sizes is observed for a-SiO_2 with increasing separation times. Since damage will take place predominantly in the region where the electronic population exceeds the critical density, the reduction in the affected area is due to the electronic decay at the wings of the Gaussian spatial distribution of the irradiation during the dark time between the pulses. The decay implies less seeding electrons, avalanche is inhibited and damage will not occur in the low energy part of the spatial profile when the next pulses are added. As expected, a diminished influence for Al_2O_3 was noted, in good agreement with the slow electron dynamics.[6] The characteristic decay time will play a role when temporally tailored excitation sequences are used for microprocessing. Smaller and controllable structures can be achieved for materials with fast electron trapping, using modulated pulses on the time scale of the electronic decay.

The production of free electrons that may ultimately lead to optical damage due to strong absorption at the critical density ($n_{cr}=1.7\times10^{21}$ cm^{-3} at 800 nm) is described by a rate equation[4,5,7] with a multiphoton term ($\sigma_6 I^6$) seeding additional avalanche ($\alpha n I$):

$$\frac{\partial n}{\partial t}=\sigma_6 I^6+\alpha n I-\frac{n}{\tau}, \tag{1}$$

where τ describes linear losses in the electron population and influences the ability to reach the critical density. Assuming a Gaussian temporal profile $I(t)=2(\ln 2/\pi)^{1/2}(F/\tau_L)\exp(-4\ln 2t^2/\tau_L^2)$ for a sufficiently short laser pulse, the following approximate relation for the electron population results:[4]

$$n(t)\cong n_0\exp\left[\int_{t_0}^{t}\left(\alpha I-\frac{1}{\tau}\right)dt\right]\cong n_0\exp\left(f\alpha F-\frac{t}{\tau}\right), \tag{2}$$

where n_0 is the number of electrons produced by multiphoton ionization of permanent defects (transient states were not considered) and valence band, and t_0 and f depend on the balance between avalanche and multiphoton processes. Since the avalanche process is seeded by the multiphoton electrons, n_0 acts as initial condition for integrating the avalanche term, setting the value for t_0. The parameter f reflects the fraction from the pulse energy used for collisional multiplication.

For a double pulse sequence of fluence F [with $I_1=I_2=(\ln 2/\pi)^{1/2}(F/\tau_L)$] with separation time $\Delta t>\tau_L$, the residual electrons $n(\Delta t)$ from the first pulse act as a seeding population for the second pulse, in addition to the multiphoton produced electrons, leading to the following expression for the electron population at the end of the sequence:

$$n\cong n_0\left[1+\exp\left(\frac{f\alpha F}{2}-\frac{\Delta t}{\tau}\right)\right]\exp\left(\frac{f'\alpha F}{2}-\frac{\tau_L}{\tau}\right), \tag{3}$$

where the characteristic influence of the decay time in the damage threshold arises. The dependence will cease when $n(\Delta t)$ becomes much lower than n_0, thus adding a negligible contribution. The solid line in Fig. 1(a) (calculated using parameters from Refs. 5 and 7: $\sigma_6=6\times10^8\text{cm}^{-3}\,\text{ps}^{-1}(\text{TW/cm}^2)^{-6}$, $\alpha=4$ cm^2/J, $\tau=60$ fs shows the estimated damage ratio for a-SiO_2 based on Eq. (1) as a function of the double pulse separation. There is a satisfactory agreement with the experimental data in view of the simplicity of the approach.

For processing, it is commonly assumed that the shortest pulses are the most beneficial, and for sapphire and fused silica this seems to be the case.[8] However, in some situations the effect is quite the opposite, when temporally modulated pulses are used. Figure 2 shows examples of microstructures induced in CaF_2 by one and three subpicosecond separated pulses per sequence for a different number of sequences per site. CaF_2 is a brittle material with high efficiency of electron trapping within 1 ps[11] and fast energy transfer to the lattice. Carrier self-trapping induces local lattice deformations and atomic displacements, softening the interaction region, and the use of modulated pulses can determine an optimum energy deposition rate. The optimum separation time is determined by a complex interplay between the energy localization and dissipation. The structures induced by triple-pulse

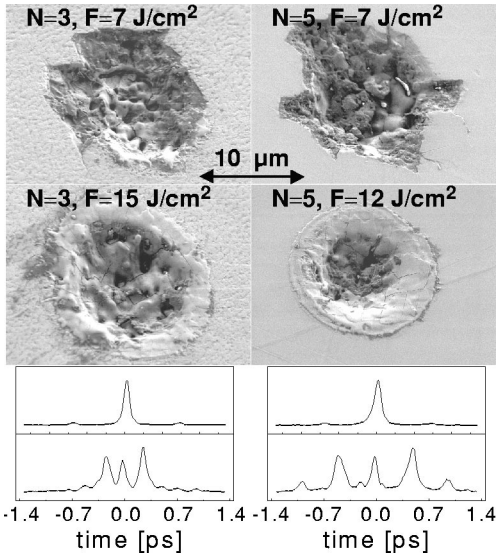

FIG. 2. Structures made on CaF_2 with single pulse (upper part) and triple-pulse sequences (bottom part) at different number of sequences per site. The fluences are increased for the triple-pulse irradiation to simulate comparable conditions for removal efficiency.

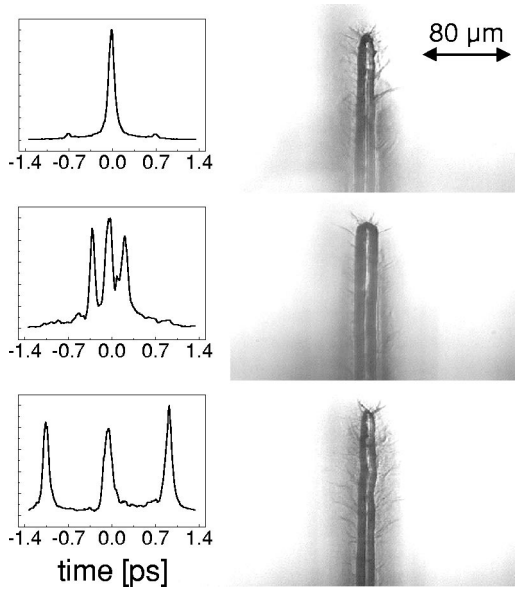

FIG. 3. Optical microscope pictures of channels drilled in a-SiO_2 with single pulses and triple-pulse sequences (shown on the left) with different separation times (0.3–1 ps) and equal fluences. Channel depth from top to bottom: 380, 310, and 400 μm.

sequences display reduced exfoliation as compared to single pulse processing and this results from a preparation of the surface induced by the first pulses in the sequence which influences the electron population and temperature control of the surface for the next pulses. The use of subpicosecond modulated pulses enlarges the processing window allowing the application of higher fluences and number of sequences per site while keeping fracturing at a reduced level. Controlled heating can be an advantage especially for brittle materials with strong electron-phonon coupling, since it provides the means for softening the surface[1,12] and therefore a relaxation of the induced stresses.

As a further proof-of-principle for the possibility of temporal optimization, we show the results for drilling deep channels ($\sim$350 μm) in a-SiO_2 with pulse trains ($\sim$600 shots, 100 J/cm^2 per sequence) with different temporal separations. As clearly seen in Fig. 3, there is an improvement in the structure when employing pulse trains as compared to the single pulse result, with an optimum for $\sim$300 fs separation. A further increase in the separation time worsens the results due to enhanced thermal stress outside the irradiated region.

This study emphasizes the benefit of using temporally designed pulses to optimize the quality of the structures induced by laser ablation. Similar efforts have been made on $\sim$ns (100 MHz) time scales, making use of cumulative heating effects at multipulse irradiation.[13,14] However, employing THz repetition rates ($\sim$subpicosecond time scales) enables controlled processing based on the synchronization between the excitation sequence and the individual picosecond response of the material.

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