Structuring of dielectric and metallic materials with ultrashort laser pulses between 20 fs and 3 ps

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ABSTRACT

Laser-micromachining of barium aluminum borosilicate glass, fused silica and stainless steel has been extended down to a pulse duration of 20 fs generated by a Ti: sapphire laser system at a wavelength of 0.8 µm. A systematic study shows that, below 100 fs, an enhanced precision and a substantial decrease of the ablation threshold fluence in comparison to pulse laser processing with pulses in the picosecond and nanosecond range could be achieved. The physical mechanism and the technical relevance of this novel microtechnology is discussed.

Keywords: ablation, laser processing, micromachining, structuring, femtosecond pulse laser, glass, silica, steel.

1. INTRODUCTION

Ultraprecision laser micromachining has excited more and more attention in different industrial fields and in medicine owing to the rapid progress in laser design capable of emitting powerful pulses with durations of less than 100 fs [1,2,3,4]. Precision and material damage achieved on the targets is determined to a major extent by the heat affected zone (HAZ) adjacent to the surface formed after each laser-induced vaporization pulse. Femtosecond laser treatment leads to HAZ's of the order of 10-100 nm compared to HAZ's in the µm range if nanosecond laser pulses are applied [5]. For a number of different materials, like metals [5], semiconductors [5,6,7], ceramics [5], inorganic dielectrics and polymers [8] and for technical [9] and biological composite materials like human corneas [10], dental and bone-like materials [5] it could be shown that 300 fs pulse laser ablation at 620 nm wavelength leads to a minimum HAZ and low ablation threshold fluence values (Table 1). Sub-picosecond-laser processing avoids any complications by plasma-light interaction (plasma shielding). It provides the possibility to use multi-photon processes which can be of importance for transparent materials [11] and to reduce the ablation threshold fluence F_{th} compared to nanosecond-laser treatment considerably [12]. Additionally, there exists no influence of the laser spot diameter on the ablation depth per pulse for constant laser fluences [13].

Since metallic and composite materials play a primary role in industrial and medical applications, the unsatisfactory structuring results with conventional nanosecond-pulse lasers call for advanced laser-technological solutions. Few reports on the pulse duration dependence of the ablation threshold fluence F_{th} in the range below 300 fs down to 100 fs have been published [14,15,16,17,18,19]. Such studies have recently been extended to pulse durations as short as 20 fs at 780 nm [20]. A reduction of the pulse duration below 100 fs yielded a further decrease of F_{th} and therefore a more efficient ablation behavior. There was conclusive experimental evidence for multiphoton ionization being dominantly responsible for optical breakdown below 100 fs in agreement with [16].

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Table 1: Ablation threshold values F_{th} for the ablation of different materials at air.
300 fs, 620 nm, 100 pulses, ~ 4 Hz, not polarized. Relative error ±30 %. [21]

Material	F_{th} [J cm^{-2}]	References
Polycrystalline gold	0.25	5,7
1 µm platinum on gold	0.1	7
Silicon(111)	0.2*	5,6
150 nm amorphous silicon on barium aluminum borosilicate glass	~0.2	7
Lime sodium glass	~1.5	8
Barium aluminum borosilicate glass (Corning® 7059)	~1.7	7,8
Fused silica (Zeiss SQ1)	~2.0	8
Polyethylene	~0.9	8
Teflon®	~1.0	8
Human cornea	0.8 (<0.5h at air); 0.45 (1-2h)	10
Human teeth	0.7-0.8 (enamel); 0.3 (dentine)	5
Bone-like material (fish scale)	0.4	5

* 1000 pulses

2. EXPERIMENTAL

We used a recently developed kHz-rate Ti:sapphire amplifier system delivering sub-20fs pulses of energies up to 0.3 mJ at 780 nm [4]. An electronic system controlling the pulse picker generated pulse bursts with a predefined number of pulses. The energy of the pulses was changed with a combination of single-order waveplate and polarizer. By detuning the prism compressor the pulses could be stretched up to 300 fs, longer pulses have been generated by inserting additional dispersive material (SF57, Schott). Pulse durations ≤100 fs have been determined with an interferometric autocorrelation technique [4]. The specimen were cleaned with aceton prior to the ablation experiments. For microstructuring, they were mounted on a motorised x-y-z-translation stage (LOT). The femtosecond-pulses were focused onto the front side of the samples with spherical mirrors with focusing distances of 15 cm and 7.5 cm, respectively. The beam diameters (laser intensity drop from maximum to $1/e^2$) have been determined with the knife-edge method. All experiments have been performed at air in a direct writing procedure.

The damage threshold was observed visually by the emergence of diffraction and scattering in the transmitted and reflected light of a He-Ne-laser beam which had been directed onto the interaction region. After laser treatment, an inspection of the ablation cavities by means of a high resolution optical microscope with a 1 µm scale (Polyvar, Reichert-Jung) has been performed. Within an experimental error <10 %, the two methods for the evaluation of F_{th} yielded the same results.

Barium aluminum borosilicate glass (1.22 mm thick, Corning® Code 7059), fused silica (0.2 mm thick, Corning® Code 7940) and stainless steel plates (0.3 mm thick, X2CrNiMo 17132) served as targets. The steel plates were cleaned after laser processing in an ultrasonic ethanol bath to remove the debris from the surface. Scanning electron micrographs of the samples (gold coated for barium aluminum borosilicate glass) were obtained by a scanning electron microscope equipped with a cold-field electron emission cathode (Hitachi S-4100) at an accelerating voltage of 20 kV.

3. RESULTS AND DISCUSSION

3.1 Dielectrics

Barium aluminum borosilicate glass and fused silica are transparent in the visible and near-infrared spectral region (Fig. 1). Therefore, it cannot be expected to observe an interaction between the materials and light, if the light wavelength is 620 nm [8] or 780 nm. But, the very high light intensities of >TW cm^{-2} enable multiphoton absorption and the processing of (1-photon) nonabsorbing materials. Additionally, incubation phenomena have to be considered [8].

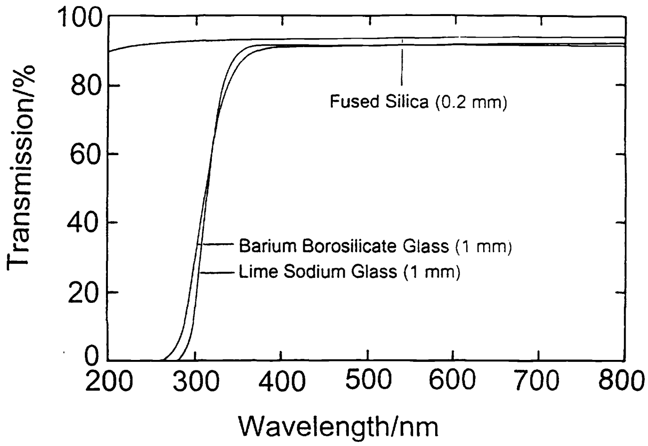

Fig. 1 Ultraviolet and visible transmission of glasses and fused silica. [8]

Ablation craters after processing with pulse durations of less than 100 fs show different quality than those obtained with longer pulses. 10 pulses with 20 fs duration e.g., resulted in smooth morphologies (Fig. 2) in contrast to ripple structures characteristic for treatment with pulses of 300 fs at comparable laser fluence [8]. This behavior cannot be explained by heat affected zone effects, but may be caused by different light penetration depths in dependence on the pulse duration. This will be described and discussed elsewhere [22].

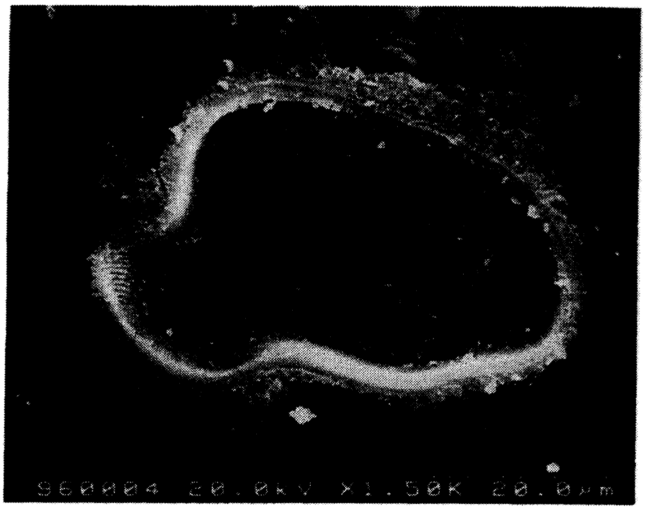

Fig. 2 Pulse laser ablation of barium aluminum borosilicate glass (Corning® 7059) at air.
780 nm, 10 pulses, 1 kHz, linearly polarized, 4 Jcm^{-2}, 20 fs.

At pulse durations above the electron-phonon relaxation time of the order of 1-10 ps, F_{th} changes with pulse duration τ according to a $\tau^{0.5}$ law [23]. Below, F_{th} of the glass and the fused silica sample show a much weaker dependence (Fig. 3).

Both materials behave similarly down to ~100 fs. Below 100 fs however, fused silica and glass deviate. The material with the smaller optical bandgap exhibits much lower ablation threshold fluences.

Using the rate-equation approximation taking into account collisional and multiphoton ionization, the evolution of the conduction-band electron density n is given by [16]

$$\partial n/\partial t = aI(t)n + P(I) \tag{1}$$

where $I(t)$ is the time(t)-dependend laser pulse intensity, a the impact ionization coefficient, and $P(I)$ the multiphoton ionization rate. For our glass sample with a bandgap of ~4 eV, 3-photon-absorption is needed for the generation of free carriers. Using the strong field Keldysh formula [24] $P(I)$ becomes $2\times10^{9} \times I^{3}$ cm^3 GW^{-3} ps^{-1}. Because of the non-availability of data needed for the calculation of a [16], we used it as a fit parameter. The damage fluence F_{th} was determined by requiring $n \approx 10^{21}$ cm^{-3}. The best fit to the experimental data was obtained with $a = 3$ cm^2 J^{-1} and is shown as a solid line in Fig. 3.

For extremely short pulses ($\tau < 30$ fs), multiphoton ionization alone provides the critical density of electrons for ablation [16]. That explains qualitatively our experimental results below 50 fs. The 3-photon-absorption process in glass gave a higher coupling efficiency and lower F_{th} than the 5-photon absorption process required in the large-bandgap material fused silica.

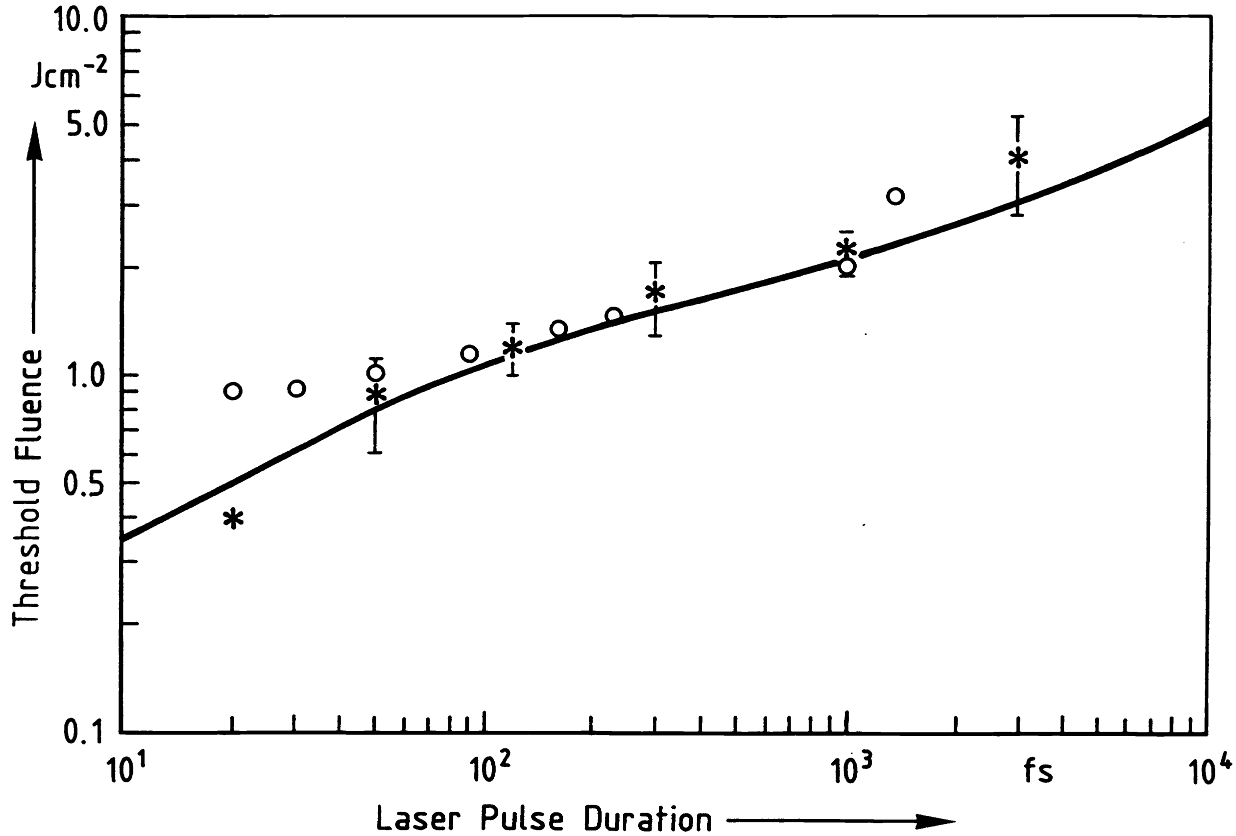

Fig. 3 Threshold fluence F_{th} vs. laser pulse duration τ for the pulse laser ablation of barium aluminum borosilicate glass (Corning® 7059, ∗) and fused silica (Corning® 7940, O) at air. 780 nm, 50 pulses, linearly polarized. The error bars are indicated for barium aluminum borosilicate glass. The solid line corresponds to the simulation for barium aluminum borosilicate glass (Eq. 1) [16].

3.2 Steel

The laser drilling of steel is a delicate task because metals show a high heat diffusion coefficient and it is difficult to avoid the formation of a large melting zone. Recently, there has been a first attempt to drill a 100 μm thick steel foil with 200 fs pulses in comparison to a 80 ps and 3.3 ns laser pulse treatment at 780 nm and a repetition rate of 10 Hz in vacuum. It could be demonstrated that the quality of the cavities in the femtosecond case was superior to that generated by ps and ns laser processing [25]. In this context, we reduced the pulse duration about one order of magnitude and performed the experiments at air which is relevant for the industrial application of the femtosecond laser technique.

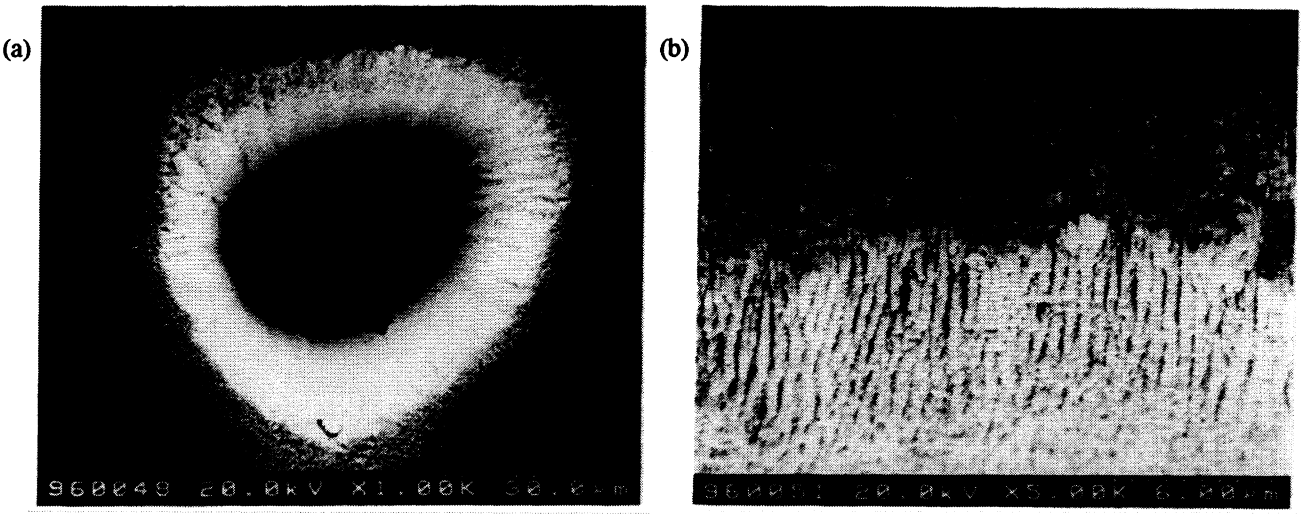

Fig. 4 Pulse laser ablation of stainless steel (X2CrNiMo 17132) at air. Front, 30 fs, 780 nm, 60000 pulses, 1 kHz, linearly polarized, 3.2 Jcm^{-2}. (a) total aspect, (b) detail.

We were able to drill holes with 30 fs pulses in a direct writing procedure at air (Fig. 4a). The cavities exhibit smooth edges and side walls with rest roughnesses <1 μm (Fig. 4b). Ripple generation is minimized compared to 300 fs laser treatment at 620 nm [5]. The redeposited debris can easily be removed by quick supersonic treatment.

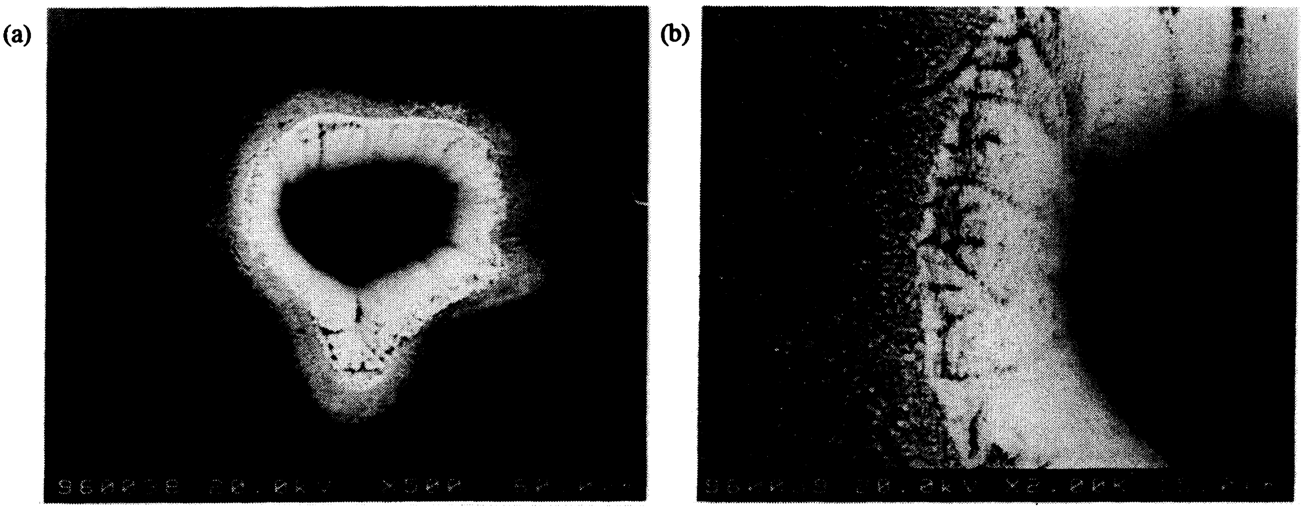

Fig. 5 Pulse laser ablation of stainless steel (X2CrNiMo 17132) at air. Front, 30 fs, 780 nm, 5000 pulses, 1 kHz, linearly polarized, 16 Jcm^{-2}. (a) total aspect, (b) detail.

The hole depicted in Fig. 4 is not drilled through the whole thickness of the stainless steel sample of 0.3 mm. With increasing fluence, it is possible to reach a throughhole (Fig. 5a). The front side shows also a smooth ablation crater with a minimum of ripples (Fig. 5b).

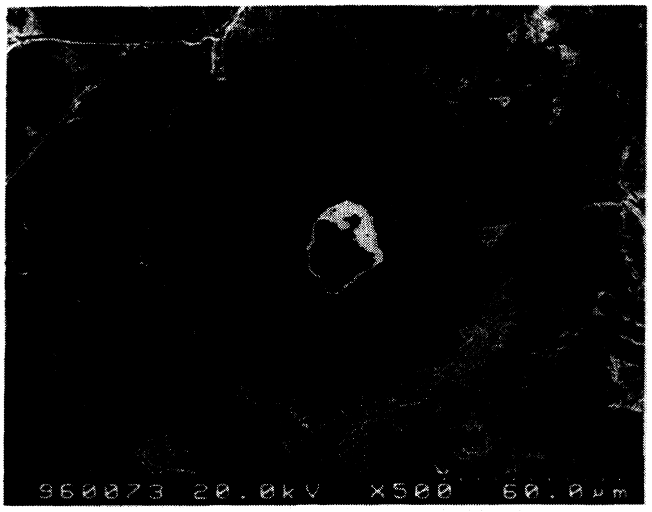

Fig. 6 Pulse laser ablation of stainless steel (X2CrNiMo 17132) at air. Backside, 30 fs, 780 nm, 5000 pulses, 1 kHz, linearly polarized, 16 Jcm^{-2}.

The backside view of the hole represented in Fig. 5 is depicted in Fig. 6. Precision parameters relevant for industrial applications are the entrance diameter (D_1), the exit diameter (D_2), the shape tolerance (ΔD = D-D'), the entrance edge radius (r_1), the exit edge radius (r_2), the flank angle (α), the inclination tolerance (u) at hole depths (d) (Fig. 7). The tangent of the flank angle is [26]:

$$\tan\alpha = \frac{D_1 - D_2}{2(d - r_1 - r_2)} \tag{2}$$

From Fig. 5a and 6 one can derive the following approximate parameters: $\Delta D < 5$ µm, $D_1 \approx 85$ µm, $D_2 \approx 25$ µm, $r_1 \approx r_2 \approx 20$ µm, d = 300 µm, $\alpha \approx 7°$, and u < 5 µm. ΔD depends very much from the spatial beam profile, and has not been optimized in this extreme short duration regime.

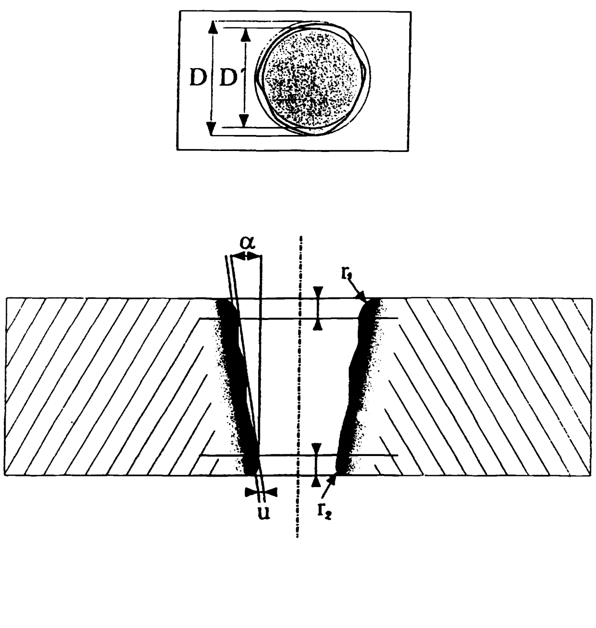

Fig. 7 Precision parameters for laser machining

4. CONCLUSIONS

The feasibility of sub-100 fs pulse laser processing of transparent and metallic materials in a direct writing procedure at air has been demonstrated. Debris can be easily removed by supersonic treatment.

Laser processing of barium aluminum borosilicate glass with pulse durations of less than 100 fs shows different quality than that obtained with longer pulses. Smooth morphologies occurred in contrast to ripple structures characteristic for treatment with pulses longer than 100 fs at comparable laser fluence. This behavior is not caused by heat affected zone effects, but by different light penetration depths in dependence on the pulse duration.

It is possible to drill throughholes in 0.3 mm thick stainless steel plates with 30 fs pulses in a direct writing procedure at air. The cavities exhibit smooth edges, a minimum of ripples and no signs of thermal damage. Precision parameters such as shape tolerance and flank angle have to be improved in further developments

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REFERENCES

1. J.-C. Diels and W. Rudolph, "Ultrashort laser pulse phenomena", chapter 5 and 6, Academic Press, San Diego, 1996.
2. M.D. Perry and G. Mourou, "Terawatt to petawatt subpicosecond lasers", *Science* **264**, pp. 917-924, 1994.
3. F. Krausz, M.E. Fermann, T. Brabec, P.F. Curley, M. Hofer, M.H. Ober, C. Spielmann, E. Wintner, and A.J. Schmidt, "Femtosecond solid-state lasers", *IEEE J. Quant. Electr.* **QE-28**, pp. 2097-2122, 1992.
4. M. Lenzner, C. Spielmann, E. Wintner, F. Krausz, and A.J. Schmidt, "Sub-20-fs, kilohertz-repetition-rate Ti:sapphire amplifier", *Opt. Lett.* **20**, pp. 1397-1399, 1995.
5. W. Kautek and J. Krüger, "Femtosecond-pulse laser ablation of metallic, semiconducting, ceramic, and biological materials", *SPIE Vol.* **2207,** pp. 600-611, 1994.
6. W. Kautek and J. Krüger, "Femtosecond-pulse laser microstructuring of semiconducting materials", *Semiconductor Processing and Characterization with Lasers, Materials Science Forum Vols.* **173-174,** Trans Tech Publications, Switzerland, pp.17-22, 1995.
7. J. Krüger and W. Kautek, "Femtosecond-pulse laser processing of metallic and semiconducting thin films", *SPIE Vol.* **2403,** pp. 436-447, 1995.
8. J. Krüger and W. Kautek, "Femtosecond-pulse visible laser processing of transparent materials", *Appl. Surf. Sci.* **96-98**, pp. 430-438, 1996.
9. J. Krüger and W. Kautek, "Femtosecond-pulse visible laser processing of fibre composite materials", *Appl. Surf. Sci.* **106**, pp. 383-389, 1996.
10. W. Kautek, S. Mitterer, J. Krüger, W. Husinsky, and G. Grabner, "Femtosecond-pulse laser ablation of human corneas", *Appl. Phys.* **A58**, pp. 513-518, 1994.
11. R. Srinivasan, E. Sutcliffe, and B. Braren, "Ablation and etching of polymethylmethacrylate by very short (160 fs) ultraviolet (308 nm) laser pulses", *Appl. Phys. Lett.* **51**, pp. 1285-1287, 1987.

12. S. Küper and M. Stuke, "Femtosecond UV excimer laser ablation", *Appl. Phys.* **B44**, pp. 199-204, 1987.

13. B. Wolff-Rottke, J. Ihlemann, H. Schmidt, and A. Scholl, "Influence of the laser-spot diameter on photo-ablation rates", *Appl. Phys.* **A60**, pp. 13-17, 1995.

14. D. Du, X. Liu, G. Korn, J. Squier, and G. Mourou, "Laser-induced breakdown by impact ionization in SiO_2 with pulse widths from 7 ns to 150 fs", *Appl. Phys. Lett.* **64**, pp. 3071-3073, 1994.

15. B.C. Stuart, M.D. Feit, A.M. Rubenchik, B.W. Shore, and M.D. Perry, "Laser-induced damage in dielectrics with nanosecond to subpicosecond pulses", *Phys. Rev. Lett.* **74**, pp. 2248-2251, 1995.

16. B.C. Stuart, M.D. Feit, S. Herman, A.M. Rubenchik, B.W. Shore, and M.D. Perry, "Optical ablation by high-power short-pulse lasers", *J. Opt. Soc. Amer.* **B13**, pp. 459-468, 1996.

17. H. Varel, D. Ashkenasi, A. Rosenfeld, R. Herrmann, F. Noack, and E.E.B. Campbell, "Laser-induced damage in SiO_2 and CaF_2 with picosecond and femtosecond laser pulses", *Appl. Phys.* **A62**, pp. 293-294, 1996.

18. C. Momma, B.N. Chichkov, S. Nolte, F. von Alvensleben, A. Tünnermann, H. Welling, and B. Wellegehausen, "Short-pulse laser ablation of solid targets", *Opt. Commun.* **129**, pp. 134-142, 1996.

19. F. H. Loesel, M.H. Niemz, J.F. Bille, and T. Juhasz, "Laser-induced optical breakdown on hard and soft tissues and its dependence on the pulse duration: Experiment and model", *IEEE J. Quant. Electr.* **QE-32**, pp. 1717-1722, 1996.

20. W. Kautek, J. Krüger, M. Lenzner, S. Sartania, C. Spielmann, and F. Krausz, "Laser ablation of dielectrics with pulse durations between 20 fs and 3 ps", *Appl. Phys. Lett.* **69**, pp. 3146-3148, 1996.

21. J. Krüger and W. Kautek, "Sub-picosecond-pulse laser machining of advanced materials", In: F. Dausinger, H.W. Bergmann, and J. Sigel (Eds.), *Proceedings of the 6th European Conference on Laser Treatment of Materials (ECLAT'96, Stuttgart)*, pp. 691-698, 1996.

22. J. Krüger, W. Kautek, M. Lenzner, S. Sartania, C. Spielmann, and F. Krausz, "Laser micromachining of barium aluminium borosilicate glass with pulse durations between 20 fs and 3 ps", in publication.

23. N. Bloembergen, "Laser-induced electric breakdown in solids", *IEEE J. Quant. Electr.* **QE-10**, pp. 375-386, 1974.

24. L.V. Keldysh, "Ionization in the field of a strong electromagnetic wave", *Sov. Phys. JETP* **20**, pp. 1307-1314, 1965.

25. B.N. Chichkov, C. Momma, S. Nolte, F. von Alvensleben, and A. Tünnermann, "Femtosecond, picosecond and nanosecond laser ablation of solids", *Appl. Phys.* **A63**, pp. 109-115, 1996.

26. German and European standard concept for pulse laser processing, 1996.