Ultra-Short Pulse Lasers for Microfabrication: A Review

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(Invited Paper)

Abstract—Ultra-short pulse lasers, generating coherent light pulses with pulse durations in the picosecond and femtosecond range, are becoming popular in precision laser microfabrication. They are benefiting not only from well-predicted laser ablation with the suppressed heat-affected zone but also by opening new processing opportunities, especially in transparent materials, due to enhanced non-linear interaction with the material. In this review, the evolution of various kinds of mode-locked lasers from a scientific toy to a robust industrial tool is reviewed. The utilization benefits of high-average-power, high-pulse-repetition-rate ultra-short pulse laser are closely related to beam shaping and manipulation techniques. Fast beam scanning with galvanometric and polygon scanners as well as multi-beam parallel processing methods were developed. Some hot applications areas are briefly described. The efficient use of photons from ultra-short pulse lasers pushed the development of optimization methods in the ablation process. Efforts toward upscaling the process by increasing the average power of mode-locked lasers made feedback to laser developers, and burst regime became a necessity as well as high-speed beam scanning devices. Unique opportunities of ultra-short pulses are successfully exploited in machining transparent materials, with glass separation being the leading application.

Index Terms—Ultra-short pulse lasers, laser applications, materials processing.

I. INTRODUCTION

LTRASHORT pulse lasers generating coherent light pulses with pulse durations in the picosecond and femtosecond range are becoming popular in precision laser microfabrication benefiting not only from well-predicted laser ablation with the suppressed heat-affected zone but also by opening new processing opportunities, especially in transparent materials due to enhanced non-linear interaction with the material. It took more than thirty years until ultra-short pulse lasers from a scientific toy working in a thermally stabilized laboratory evolved into a robust tool for the 24/7 operation at a factory floor.

Increased pulse repetition rate from 1 kHz to tens of MHz and even GHz of industrial-grade lasers pushed progressing in fast beam scanning with galvanometric and polygon scanners. Furthermore, upscaling in pulse energy of the ultra-short pulse

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lasers lead to innovations in multi-beam processing methods with the highly parallelized material process with a single laser, including utilization of laser beam interference.

Together with the evolution of the picosecond and femtosecond lasers, significant efforts have been made in laser process development, utilizing unique properties of the coherent light sources with extremely short pulse duration. Some hot applications areas are briefly described.

The efficient use of photons from ultra-short pulse lasers pushed the development of optimization methods in the ablation process. Efforts toward upscaling the process by increasing the average power of mode-locked lasers made feedback to laser developers, and burst regime became a necessity as well as high-speed beam scanning devices. Unique opportunities of ultra-short pulses are successfully exploited in machining transparent materials, with glass separation being the leading application.

II. ULTRA-SHORT PULSE LASERS

Ultra-short pulse (USP) lasers are ones working in a modelocking regime with pulse duration from tens of picoseconds down to femtoseconds when spontaneously emitted photons are synchronized during their travel inside the laser cavity. There is a variety of methods to generate ultra-short light pulses, but stable pulse trains from diode-pumped solid-state lasers are provided by Kerr-lens, saturable semiconductor absorber mode-locking or in a fiber laser.

Industrial-grade ultra-short pulse lasers, solid-state or fiber-based, consists of a master oscillator, power amplifier and optional stretcher/compressor to get sub-picosecond pulses.

A. Master Oscillators for USP Lasers

The master oscillator is the heart of the USP laser, where coherent light pulses are synchronized and temporally shaped. The resonator cavity possesses a special mean for mode-locking. A stimulus to make robust USP lasers, running at a high pulse repetition rate of 100s kHz and beyond, was introducing a low-loss fast intracavity semiconductor Fabry-Perot saturable absorber, known as SESAM, to start and sustain stable passive mode-locking [1]–[3]. Parameters of SESAM could be easily adopted to particular laser design by variation in the multi-layered structure of semiconducting materials. However,

despite significant progress in design and manufacturing technologies [4], the resistance of semiconductor structures to laser radiation is limited, leading to SESAM degradation during the operation [5].

Another approach used in the USP laser is the Kerr-lens mode-locking [6], [7] via the nonlinear optical Kerr effect, based on that, the refractive index of a medium is dependent on the electromagnetic field strength. This method allows the generation of light pulses with duration down to a few femtoseconds. For example, pulses shorter than two optical cycles with bandwidths over 400 nm have been generated from a Kerr-lens mode-locked Ti:sapphire laser with a repetition rate of 90 MHz [7].

Both those approaches for mode-locking do not well fit with the all-in-fiber design of modern fiber lasers. Fiber-based USP lasers are attractive because of their compactness, alignment-free set-ups and low cost. Regelskis *et al.* demonstrated a Yb-doped fiber ultra-short pulse generator based on self-phase modulation and alternating spectral filtering, operating at a wavelength of 1060 nm and providing a stable ultra-short pulse train [8]. High intensities confined within a fiber core initiate nonlinear phenomena with the dynamic evolution of the gain spectrum. The so-called Mamyshev oscillator scheme is becoming popular in fiber-based USP lasers [9], [10] with a potential to get high-energy, sub-30 fs pulses from a compact fiber source.

B. Amplified Ultra-Short-Pulse Laser Systems

The pulse energy available from the oscillator is too low for most applications in material processing, and additional stages for their amplification are integrated to make the real industrial tool. Therefore, high power USP lasers are assembled in MOPA (Master Oscillator Power Amplifier) design with regenerative multi-pass amplifiers and/or linear power boosters. High intensity inside a laser gain media may damage it. D. Strickland and G. Mourou [11] proposed temporal stretching of laser pulses before amplification and subsequent their compression. The method called Chirped Pulse Amplification (CPA) is a core of all high-pulse-energy femtosecond lasers now. Stretching and compression are realized with pairs of diffraction gratings [12], fiber Bragg gratings [13], [14] and volume Bragg gratings [15].

Solid-state lasers with a rod-shaped active media still remain the main type of ultra-short pulse lasers at a power level below 100 W. Due to the broader gain spectrum, Yb-based crystals are used in diode-pumped femtosecond lasers, while Nd-doped crystals are used in the lasers with a pulse duration above 10 ps. Scaling the average power of rod-type lasers is limited by heat, dissipated in solid-state media, which is a byproduct of the lasing and significantly impacts lasing performance [16]. Non-uniformly distributed heat within the bulky rod modifies refractive index (thermo-lens), induces stress and physical deformation of the material, which leads to a wave-front distortion. Fiber or thin-disk geometry of the active laser media facilitates easier removal of heat, significantly reducing the detrimental thermo-lensing effect.

The advent of fiber lasers in the laser processing field, especially in macro-processing with continuous-wave (CW) kW-class devices, pushed efforts in the development of robust USP

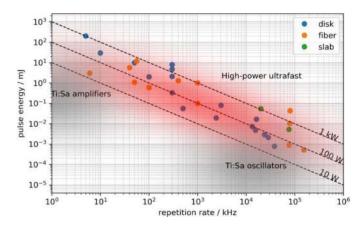


Fig. 1. Pulse energy versus repetition rate of representative state-of-the-art ultrafast lasers based on different geometries, both amplifiers and oscillators [24]

sources of coherent radiation. Fiber lasers are associated with high average powers and very high beam qualities needed for many industrial applications [17]. Benefiting from excellent thermal management in the active media, beam quality and single-mode operation in 10 kW range, mode-locked operation of fiber lasers possess challenges due to extremely high intensities inside the core of the fiber. The major limitations are imposed by fiber nonlinearities, optical damage, transverse modal in-stabilities and photodarkening [18]. To make stable, practical, and maintenance-free lasers, new types of saturable absorbers with a nanocarbon material and ultra-short pulse compression techniques [19] and scaling the pulse energy and peak power [18] were developed. The performance of ultrafast fiber lasers is well defined by the laser parameters, including repetition rate, spectral bandwidth, pulse duration, pulse energy and average power [20].

In contrast to the long active fiber, a thin disk is only a few 100's micrometers thick and requires multi-pass travel through the gain media of the laser light [21], [22]. Efficient heat removal through the backside nearly eliminates the thermo-lensing problems. The results for amplification of ultra-short pulses to average output powers over 200 W in sub-picosecond regime demonstrate the potential of the thin-disk laser design. The mode-locked thin-disk laser generating 80 μ J of pulse energy 242 W of average output power with a pulse duration of 1.07 ps without additional amplification has been demonstrated in [23].

In recent years, significant progress in thin-disk ultra-short pulse lasers was achieved, including novel gain materials with broadband gain, efficient external pulse compression using hollow-core fibers, and multi-pass cells (Fig. 1) [24]. Further scaling of average power requires multiplexing the gain amplification elements to spread the thermal load over several disks.

Thin-disk lasers are indispensable in industrial applications. They represent a unique class of lasers that provides kW output power with excellent beam quality, long-term stability, thermal management, and power scalability. Due to multi-pass configuration, thin-disk lasers are engineering masterpieces with sophisticated multi-mirror arrangements, and the reduced complexity of the laser is highly beneficial. A monolithic pump design for

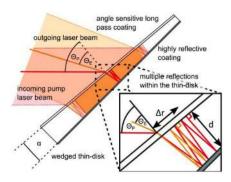


Fig. 2. Close-up sketch of wedged thin-disk geometry. Indicated are the wedge angle α , the two incidence angles for pump (Θ_P) and lasing (Θ_E) radiation, as well as the local thickness d and beam displacement Δr . The upper surface is coated as a long pass, while the bottom (back) side is coated with a wide-angle, highly reflective coating glued onto a diamond heat sink [25].

thin-disk lasers and amplifiers is demonstrated in [25]. The thin disk is replaced by a thin, wedged gain medium acting as a wedged optical trap (Fig. 2). The efficient coupling of the pump and laser radiation and 890 W of CW output power with the optical-to-optical efficiency of 50% has been achieved.

C. Power Upscaling of USP Laser to kW Level

A wide field of materials processing applications requires ultra-short-pulse lasers delivering both high average and peak powers at a high repetition rate and efficiency. Most ultra-short pulse lasers utilized in the industry today are in the power range of 10 W to 100 W, when it is still possible to control irradiation of the material by a single laser spot. To be more competitive with alternative technologies, high power lasers with an average power reaching 1 kW and more are on development.

A thin-disk multi-pass amplifier for ultra-short laser pulses delivering an average output power of 1105 W, the pulse energy of 1.38 mJ at a duration of 7.3 ps was reported in [26]. The beam quality better than M2 = 1.25 was maintained at that power level. Potentials of power scaling using the thin-disk type lasers were shown by Dietz *et al.* [27]. An ultra-short-pulse Yb-doped thin-disk laser system without chirped-pulse amplification in the multi-pass set-up provided a maximum output power of 1.9 kW at a repetition rate of 400 kHz. The total energy in the four-pulse burst regime was as high as 46.7 mJ at a repetition rate of 25 kHz. A thin-disk laser with output power exceeding 10 kW of and high beam quality and consisting of two thin-disk laser oscillators and a thin-disk multi-pass amplifier system was presented in [28]. Those examples show the great potential of thin-disk lasers to scale up their power in a model-locked regime.

The slab geometry of the active media allows heat removal through a side wall for the laser crystal. Therefore, this approach is also utilized in upscaling the average power of USP lasers. One successful example is the Innoslab amplifier comprising a diode-laser end-pumped thin slab crystal and a folded single-pass optical amplification path [29]. Up to 5 kW average power, a few Joule pulse energy are possible from the Innoslab design. The complete laser is a complex system and includes a fiber MOPA, rod-based amplifier and few stages of Innoslab to get to

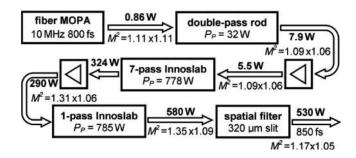


Fig. 3. Spatially filtered Innoslab MOPA system [29].

more than 0.5 kW power level with excellent beam quality with $M^2 < 1.2$ (Fig. 3).

Scaling average power or pulse energy in ultra-short-pulse fiber lasers is limited, even utilizing large-mode-area (LMA) or photonic-crystal (PCF) fibers. The limitations come from nonlinearities induced in the core disturbing beam quality and spectrum. Parallel amplification and combining multiple beams in a single high-brightness beam is a scalable laser architectural approach. Coherent beam combining (CBC) of 61 femtosecond fiber chirped-pulse amplifiers with the efficiency of $\sim 50\%$ is demonstrated [30]. The first CBC femtosecond laser delivering 10.4 kW average output power was demonstrated in [31] by a coherent combining of 12 step-index fiber amplifiers.

D. GHz Laser Sources for Material Processing

A new branch of USP laser sources approaching in recent years is generating a burst of pulses with temporal separation between pulses in nanosecond and picosecond time frames. Ultrafast lasers operating at high repetition rates, particularly the GHz range, enable new potentials in material processing, particularly accessing the recently demonstrated ablation-cooled regime [32]. Ultra-high pulse repetition rate is a way of upscaling the process efficiency. The laser could be implemented in the burst mode to access GHz repetition rates and microjoule-level pulse energies without requiring kilo-watts of average power [33]. The 2.65 GHz repetition rate bursts of ultra-short laser pulses containing a desirable number of pulses within a burst with identical pulse separation and adjustable amplitude were demonstrated in an active fiber loop [34]. A burst mode all-fiber laser system operating at an intra-burst repetition rate of up to 1.2 GHz with pulse duration <500 fs and average power higher than 100 W [35]. That all-fiber laser could be a promising tool for micromachining applications requiring fs pulses with both high average power and high repetition rate.

An industrial-grade 100 W femtosecond GHz-burst laser has been developed, delivering total burst energy up to 1 mJ at 100 kHz, with an adjustable number of pulses per burst. The intra-burst repetition rate could be varied between 0.88 and 3.52 GHz, with the maximum number of pulses per burst of 3200 [36], [37]. The solid-state laser system working in a multi-burst regime with a controllable number of pulses and tunable pulse-to-pulse time spacing ranging from 200 ps to 16 ns is presented in [38], [39]. The laser generates pulse bursts of the GHz to MHz repetition rate, with the burst repetition rate

ranging from 100 kHz to a single shot and a constant duration of 300 fs.

The GHz intra-burst repetition rate in UV spectral range was achieved based on the fourth-harmonic generation of an electro-optic comb operating in a burst mode which permits flexible repetition rate at the GHz level [40]. An ultra-short pulse laser combined with an external array of birefringent crystals was applied to generate near-THz bursts of single-picosecond pulses at variable delays between consecutive pulses [41].

III. LASER PROCESSING SYSTEMS

During the recent ten years, significant progress was made in ultra-short pulse lasers, increasing their average power, pulse repetition rate, stability, robustness and flexibility. The processing quality, in general, meets the demands of many applications. However, processing with USP lasers still lacks productivity for industrially relevant cases. The average power of a pulsed laser source is the product of pulse energy and repetition rate. There are two approaches for upscaling USP processes [42], [43]:

- 1) by increasing the pulse energy.
- 2) by increasing the pulse repetition rate.

As the productivity and throughput of material processing using USP lasers should be competitive with alternative technologies, tremendous efforts were also added in progressing of the laser energy management outside of the lasers. A variety of mechanical and optical systems are used to control the position, direction and shape of the laser beam over the workpiece.

Material processing utilizing ultra-short laser pulses enables precise fabrication with a negligible thermal load. However, even with ultra-short laser pulses, not the whole laser energy absorbed by the target is removed by ablation products. The residual energy is converted to heat which accumulates pulse-by-pulse or scan-by-scan. Heat accumulation occurs due to very short time separation between pulses impinging the same spot on the work-piece [42], [44]. That is a severe problem when high repetition rate (>100s kHz) lasers are applied. Fast motion of the laser beam is required to keep beam overlap within acceptable limits. The multi-pulse irradiation with the laser beam significantly lowered the ablation threshold. It led to a relative increase in the ablation rate at the higher repetition rate, but plasma shielding limits the processing efficiency of metals with the high-power picosecond lasers [42].

Temperature rise on the workpiece surface has a severe adverse impact on the ablation quality in USP laser processing of metals [45]. However, heat accumulation was identified as the main factor responsible for the enhanced ablation efficiency preserving excellent quality during the burst mode processing of diamond-like nanocomposite films with femtosecond laser [46].

If the laser is emitting high pulse energy, the processing speed upscaling could be realized by splitting the pulse energy of the incoming beam. Therefore, this kind of lasers is usually applied in a multi-beam processing approach.

A. Galvanometric and Polygon Scanners

The most robust and convenient fast positioning of a laser beam is achieved utilizing a galvanometric scanner with nearly

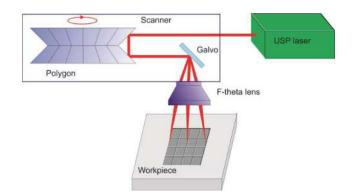


Fig. 4. Combination of polygon scanner with a galvanometric scanner for fast 2D laser beam deflection. Adopted from [55] with permission of JLMN.

inertia-less tilting of two mirrors in perpendicular directions. Modern galvoscanners with an f-theta objective of 160 mm focal length move the laser beam at the speed of 20 m/s in the 100 mm x 100 mm field [47], [48]. Synchronization of laser pulsing with the motion of the laser beam is becoming challenging at such speed. As most USP lasers are self-mode-locked, external synchronization is not feasible. External synchronization of galvoscanner from the USP laser clock was proposed by B. Neuenschwander et al. [49]. The precision of pulsing is determined by the pulse repetition rate of the oscillator (seeder), which usually is in the range of 30-100 MHz. The synchronization facilitates the positioning of small ablation craters from separate laser pulses with the precision of about 1 μ m relative to each other at a high laser-beam translation speed. The outcome is a smooth surface at the edges of ablated cavities and a high taper angle of them. The approach was extended for laser engraving of printing and embossing tools with a rotation symmetry with a precision of 1 μ m [50].

Polygon scanners, broadly used in imaging and bar-code reading, are still new in the material processing field. They are able to move laser beam at a speed of 100–1000 m/s on the workpiece surface [51]–[54]. However, synchronization of USP laser pulsing with highly stable rotation of the polygon is more challenging. Intelligent control systems and software are required. The spatial resolution and precision of laser treatment are limited by the pulse repetition rate of the oscillator. A synchronized high-speed beam deflection using polygon scanners permits overcoming plasma shielding and heat accumulation limitations utilizing multi-MHz USP lasers. The pulse overlapping is reduced due to the very fast scanning speeds, and high-average-power lasers can be used very efficiently in high-throughput material processing.

Combining a polygon scanner with a single-axis galvoscanner, a fast two-dimensional scanner was developed (Fig. 4) and validated with a 1 kW laser in multi-pass drilling in stainless steel and silicon [55]. The distribution of consecutive laser pulses over the whole laser-treated area decoupled the heat-accumulation and plasma-shielding effects.

The new type of resonant scanner with sophisticated control to compensate the oscillation nonlinearities and an additional galvanometer scanner for processing two-dimensional fields was

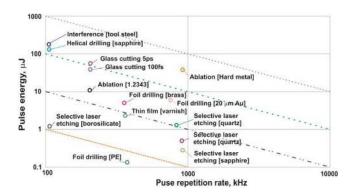


Fig. 5. Process landscape for typical USP laser applications based on pulse energy and repetition rate. Diagonal lines of equal average power are marked. Drilling of metallic foils and thin-film structuring uses comparable low power <10W. Only some applications, as glass cutting and interference patterning, utilize higher power and pulse energy. Adopted from [67] with permission of JLMN.

proposed in [56]. The scanner was applied with a USP laser running at a 5-10 MHz repetition rate to write 1.6 million dots per second with a positioning reproducibility of 1 μ m.

The dynamic performance of galvoscanners in a small area could be extended by combining an acoustic-optical deflector (AOD) with a galvanometric scanner [57]. AOD is able to deflect the laser beam pulse by pulse within its scanning field and modulate the beam intensity simultaneously. As no mechanical parts are in AOD, it is also used to compensate vibrations and overshoots of fast mirror rotations resulting in process time reduction and processing quality improvement.

B. Laser Beam Shaping

Most of the lasers are emitting a beam with a Gaussian beam profile. The intensity in the center of the beam is high, and wings of low intensity spread for long distances. Such spatial energy distribution is not beneficial for many applications, especially in thin-film processing. Laser beam shaping and homogenization techniques could optimize the shape for a large number of laser-material processing applications. Diffractive optical elements (DOE) are able to transform the circular Gaussian beam into a rectangular-shaped flat-top one, where intensity is preserved in the significant part of the beam diameter providing the process-adapted laser beam shaping [58].

Diffractive optical elements offer a wide range of beam profiles, but their trade-offs result in reduced brightness and efficiency. With advanced diamond tuning technologies, free-form optics is becoming popular, including beam shaping applications [59]. Free-form refractive optics offer a wide range of profile options for high-power laser beam shapers. They can generate application-specific beam profiles with high efficiency.

Ultra-short laser pulses are complex with a broad spectrum and alteration during propagation through media, like lenses. A spatiotemporal focusing of femtosecond pulses is applied to concentrate laser pulse energy in time and space for more predictive and efficient material processing [60], [61].

A novel concept for dynamic focus shaping based on highly-efficient (\sim 90%) coherent beam combining with micro-lens arrays as the combining element was presented in [62].

C. Beam Shaping With Spatial Light Modulators

A flexible option for laser beam shaping is the use of spatial light modulators (SLM) based on a pixelated device with electroswitchable liquid crystals. Computer-generated holograms are transferred to SLM control electronics to set a phase or amplitude mask for the laser beam. SLM is applied with femtosecond lasers to produce multiple diffractive beams for parallel processing, gaining throughput of a high precision micro-structuring of silicon and titanium alloy by more than ten times [63], [64]. Optical SLM efficiency into first-order beams was $\sim 50\%$, and a 50 Hz refresh rate limited the real-time processing. The use of SLM with high power USP lasers (>100 W) require cooling of the device [65].

Usually, SLM is used to split the original laser beam to the desired number of beams with equal intensity. Separation of the laser beam to 400 beams with individually controlled intensity was presented in [66]. The technique was validated by femtosecond laser ablation of grayscale pictures individually set intensity per pixel of a corresponding beam.

D. Multi-Beam Systems

Using high power USP lasers working at a high pulse repetition rate in the MHz region could cause thermal management issues like overheating and melt formation, reducing the ablation quality. A high ablation quality could only be achieved if all process parameters are carefully matched. High beam deflection speed using advances galvanometric or polygon scanners not always provide solutions in precise microfabrication. The use of multiple laser beams provides the best and most versatile high power ablation solution [67].

Massively parallel processing could be arranged with high power USP lasers. The beam emitted by the laser is transformed into an arrangement of multiple laser beams with specific intensity distributions [68]. The main challenge in using the multi-beam optical system is the distortion of the spot-array on the workpiece. Active and passive concepts to reduce or eliminate distortion in multi-beam systems are implemented.

A fully reflective 3x3 beam splitter compatible with high power up to 300 W from a 500 fs-pulse laser is presented in [69]. The process was validated in ablating craters with multiple Gaussian beams with $15 \mu m$ diameter and $300 \mu m$ pitch.

E. Approaches for Laser Processing of Complex 3D Surfaces

Ultra-short pulse lasers are well defined for precise fabrication, including ablation of complex cavities in molds, micro- and nanotexturing 3D surfaces. Laser treatment of freeform surfaces is feasible with laser processing systems having a large number of degrees of freedom to position focused laser beam relative to the workpiece [70]. The algorithm incorporates the tessellation of a freeform surface into laser processing fields, utilizing the capabilities of high dynamics 3D galvoscanners. Limitations

of the particular laser process to focal off-set and the angle of incidence to the process performance should be considered.

F. DLIP -Direct Laser Interference Patterning

Periodic structures are of increasing interest in many fields, especially for surface functionalization. Interference of coherent laser beams produces periodic laser intensity patterns, which could be easily imprinted on workpiece surface or ablated in thin films [71]. Fabrication time reduction up to a few orders of magnitude using DLIP compared to direct laser writing was evaluated.

Various set-ups are utilized for direct laser interference patterning (DLIP). Beam splitters and mirrors are usually used in two-beam interference set-ups. In general, it has a lot of freedom to manipulate the angle of incidence and beams orientation. However, such an arrangement for three beams is a quite complex and bulky three-dimensional optomechanical system [72]. Diffractive optical elements are applied in multi-beam (3, 4, 6 beams) interference systems to split the initial laser beam and an optical system to combine the beams consisting of a confocal 4f lens system [73], [74], a prism and lens [75]–[77], or mirrors.

The process scaling is straightforward by increasing pulse energy. As energy density (fluence) required for ablation, most of the materials is close to 1 J/cm², a few squared millimeters of the surface are treated by a single laser pulse. The period of the structure depends on the number of interfering beams, the angle between them and the laser wavelength:

$$\Lambda_4 = \frac{\lambda}{\sqrt{2} \cdot \sin \Theta} \tag{1}$$

where Λ_4 is the period of a four-beam interference pattern, θ is the half-angle between opposite beams, and λ is the wavelength of laser radiation. The period could be easily downscaled to the sub-micrometer range using ultra-short pulse lasers [78]. Flexibility in periodic pattern formation has been shown by the adjustment polarization of each interfering beam (Fig. 6) [79].

Application field of DLIP in laser surface texturing is expanding rapidly from surface wettability control [80], [81], to wear resistance [82], ice-repellent surfaces [83], [84], anti-counterfeit [85], frequency-selective surfaces [73], [79], texturing of hotembossing tools [86], antibacterial surfaces [87], [88].

DLIP is combined with direct laser writing (DLW) or laser-induced periodical surface structures (LIPSS) to get hierarchical micro- and nanostructures, which are responsible for stable long-term performance of the functionalized surfaces [77], [81], [84].

IV. APPLICATION OF ULTRA-SHORT PULSE LASERS IN MICROFABRICATION

Highly concentrated in time and space energy of ultra-short laser pulse from one side leads to extremely high intensities of light irradiation able to evaporate any material. From another side, the pulse energy can be precisely adjusted, leaving invisible but real modifications inside of transparent materials. Adding that USP lasers became reliable, industrial-grade tools approaching kW power range, we have a broad field of diverse USP lasers applications in material processing. The most substantial

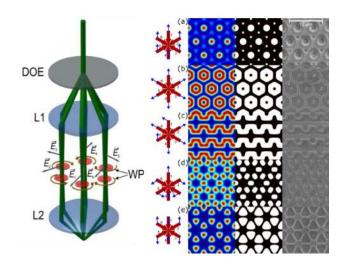


Fig. 6. (a) DLIP set-up. The laser beam is split into six beams using diffractive optical element (DOE), and beams are combined on the sample using the two-lens confocal imaging system. (b) Arrangement of laser beam polarizations, simulated periodic intensity profile, its cross-section at selected intensity level corresponding to the metal film ablation threshold and SEM images of the structures ablated in thin Cr film.

benefits are in precise micro- and nanofabrication and unique opportunities in the localized modification of transparent materials, still not fully discovered and being at the very initial stage of their exploitation. The ablation process could be easily controlled as the material is removed in small quantities. However, a high number of laser pulses is required to ablate a significant amount of the material.

A. High-Throughput and Precision Ablation of Metals

Ultra-short pulse lasers remain complex electro-optical-mechanical systems, and photons emitted by the lasers are still expensive. Tailored utilization of each photon is required for cost-effective introduction into industry and competition with alternative technologies in material processing. Photon energy is absorbed by electrons and transferred to ions in the material. Properties of the material and its response to laser excitation are changing instantaneously. Too little energy does not create permanent modification like an ablation crater. Too much energy directed to the target heats the material and ignites plasma which creates detrimental effects on the workpiece. The idea of the existence of the optimal laser irradiation condition for material removal (ablation) was proposed by Furmanski *et al.* in 2007 [89] and later developed by us [42], [44] and B. Neuenschwander group [90], [91].

As the penetration of light into non-transparent materials is limited by a high absorption coefficient, the amount of material affected by single-pulse irradiation is small. The volume of the laser-ablated crater can be found by integrating the crater profile [89], [44]:

$$V = \frac{\pi w_0^2 \delta}{4} \left(\ln \frac{F_0}{F_{th}} \right)^2 \tag{2}$$

where w_0 is the beam diameter on the surface at the intensity $1/e^2$ level; δ is the effective absorption depth; F_{th} is the ablation

threshold fluence, and F_0 is the laser fluence in the center of the Gaussian beam.

The volume of ablated material is maximal when the laser beam is focused to an optimal spot, which depends on the pulse energy and the ablation threshold. The most efficient material removal takes place when the laser fluence in the beam center is equal to:

$$F_{0\max} = e^2 F_{th} \approx 7.4 F_{th} \tag{3}$$

The model is valid for fluences close to the ablation threshold as the energy coupling might be affected by the ablation products. It was validated by may research groups on various metals: stainless steel [45], [53], [92]–[94], copper [53], [92], [95], [96]–[98], aluminum [53], [52].

The material removal rate scales directly with the average power of the laser and could be boosted by increasing the pulse repetition rate. However, residual energy, not removed by ablation products, heats the target [91]. Ramping the scanning speed with polygons and utilizing the interlaced mode facilitate temporal and spatial separation between laser pulses and prevent the target from heating to melting temperature [93], [99]. Lasers with a power of 100 W at tens of MHz could be used for high-quality ablation at a 100 m/s scanning speed. The metal removal rate in the range of 40 mm³/min was achieved with average laser powers above 300 W.

Heat accumulation is a significant challenge for materials processing with high average power pulsed lasers and repetitive processing. The minimum scanning speed yielding a high-quality surface was identified as an essential process parameter [45]. Working at the optimum point with the highest efficiency and keeping the high machining quality sets the process strategy, which can only be fulfilled by either very high marking speed or a large focal spot. High-pulse repetition frequency USP lasers are used in conjunction with high-performance galvanometer scanners and two-axis polygon scanner systems to benefit from optimal laser ablation [53].

The optimal ablation regime was achieved by adjusting laser fluence with a focus off-set, varying not the pulse energy but beam diameter on the surface to utilize the full power of 40 W from the USP laser [96]. Moreover, a parameter window for the most efficient ablation corresponded with the highest surface quality (Fig. 7).

A semi-empirical model was developed to predict optimal process parameters for efficient ablation [100]. The model considers the reduction in the ablation threshold by accumulation effects and the saturation of the ablation depth with the increasing number of pulses per spot. The model was validated in the ablation of rectangular cavities in metals and applied in a high-speed fabrication of mimicking bio-inspired functional surfaces.

Managing laser beam deflection at tens of MHz repetition rate still remains challenging. Advanced USP lasers are able to deliver high pulse energy as a way to ramp the average power. The approach to be close to the optimal ablation regime is to split laser pulse temporally into a burst of pulses with lower energy with time separation between sub-pulses corresponding to the MHz or even GHz repetition rate, while the burst repetition rate

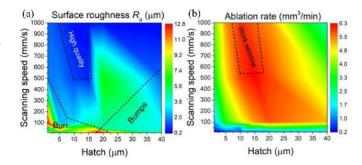


Fig. 7. Effect of the scanning speed and hatch between scanned lines to laser ablation performance: (a) dependence of the cavity bottom surface roughness after a single scan on the scanning speed and hatch distance. (b) Dependence of the ablation rate on the scanning speed and hatch distance evaluated from a multiple-scan ablation to reach a depth of 50 to 100 μ m. Reprinted from [96], with permission from Elsevier.

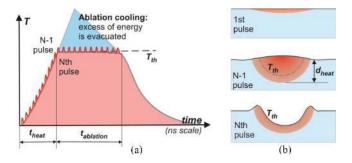


Fig. 8. The two-stage mechanism for GHz ablation: (a) Heating by accumulation effects during $t_{\rm heat},$ followed by efficient ablation of hot surface layer during $t_{\rm ablation}.$ (b) Crater generation mechanism: local temperature increase by heat accumulation triggers a massive material ejection when achieving the critical temperature $T_{\rm th};$ $d_{\rm heat}$ - heat diffusion length within $t_{\rm heat}.$ Adapted with permission from [102] © The Optical Society.

being below 1 MHz. Stimulus to research in this area and new laser development was given by a paper of C. Kerse *et al.* [32] on so-called "ablation-cooled material removal". According to their results, at the GHz repetition rate, bursts of laser pulses ablate the target material before the residual heat deposited by previous pulses diffuses away from the processing region. A very high repetition rate heats the materials in the irradiation zone and reduces the pulse energy needed for the ablation, increasing the removal rate by order of magnitude. The two-temperature simulations for the copper irradiated by 1 ps pulses at 1030 nm predict an enhancement in the ablation depth 4.2 times compared to a single pulse when an optimized 128 sub-pulse burst at 10 GHz is utilized [101]. Due to the heat accumulation from the preceding pulses, the multi-pulse laser ablation could be advantageous over a single-pulse one.

The existence of the minimum number of pulses in the burst to get an increase in the material removal rate was experimentally shown in [102] by punching (drilling holes) in copper. A heat-accumulation-based incubation is necessary for enhanced material ablation (Fig. 8).

The ablation efficiency for copper, aluminum, and stainless steel was comparable to the case of single nanosecond pulses but with the usual quality of femtosecond processing [36], [37]. L. Zhibin *et al.* [103] found that benefit up to $\sim 3.5x$ is possible only

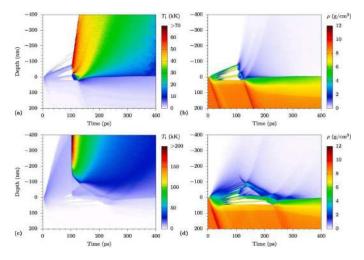


Fig. 9. Heatmaps showing changes in the ion temperature and density of a copper target when irradiated with double pulses at fluences of 1.5 J/cm², and 2.5 J/cm² for each pulse, corresponding to the top row (a) and (b), bottom row (c) and (d) of the figure, respectively. The time delay between the two pulses is 100 ps. The left column (a) and (c) and right column (b) and (d) in the figure show the ion temperature heatmaps and density heatmaps, accordingly. The white regions in the ion temperature heatmaps represent either regions with extremely low density or areas of relatively low ion temperature. Reprinted from [106], with permission from Elsevier.

in drilling holes with 864 MHz pulse bursts in copper. However, the efficiency for milling was more than two times lower than an optimized non-burst process. An optimized three-pulse burst at $80\,\mathrm{MHz}$ was $\sim 20\%$ more efficient as a single pulse [103], [104]. The plasma shielding effects and redeposition of material during the processing of metals with bursts of ultra-short laser pulses are responsible for the significant difference in the ablation efficiency for copper utilizing 1, 2 or 3 laser pulses in the burst [105]. Adding a third pulse to the burst leads to more material ablation than a single pulse. Redeposition of the material by the second pulse was confidently shown by atomistic simulation of ultra-short pulsed laser ablation of metals with single and double pulses [106]. Characteristics of the redeposition process are highly dependent on the fluence (Fig. 9).

Research on the use of short-burst lasers with a broad spectrum of parameters exploded during the recent year, supported by new developments in USP lasers with a considerable variety of burst modes available. The specific material removal rate drops down to less than 10% for the metals and 25% for silicon compared to single pulses when the number of pulses per burst is increased up to 25 pulses with a time separation of 180 ps [107]. However, the specific removal rate for soda-lime glass and sapphire increased by a factor of 2.3 and 6, respectively [108]. Both cases show the effects of shielding and heat accumulation on the ablation process.

The enhanced ablation efficiency due to heat accumulation was observed in the processing of diamond-like nanocomposite (DLN) a-C:H:Si:O films with a femtosecond laser in the burst mode [46]. The high-quality high-throughput laser milling of silicon of 230 mm³/min was achieved with a sub-ps laser delivering more than 1 kW of average laser power working in the five-pulse

burst regime and limiting the peak fluence to approximately 0.7J/cm² by expanding the beam diameter [109].

Combined GHz and MHz burst mode (bi-burst regime) were applied to process copper and steel [39]. The high-efficient USP laser processing was achieved at the fundamental 1030 nm wavelength. In the case of the MHz burst milling and drilling in copper, the ablation efficiency was highly dependent on the odd and even number of pulses per burst. The GHz processing was inefficient in milling and drilling copper and steel compared to single-pulse processing. The percussion drilling throughput can be of an order of magnitude higher when using the MHz burst mode than the optimized conventional kHz case for Invar [38].

Those controversial results show that GHz and burst processing efficiency are highly material dependent. Therefore, the process optimization should be tailored for a specific application (drilling, milling) and thermo-physical properties of materials. The most recent review on burst laser processing is in [110].

B. Surface Texturing

Surface texturing is an emerging field of applications for USP lasers providing high flexibility in functionalizing material interfaces based on optical, mechanical, or chemical properties. The laser techniques depend on the required texture geometry and characteristic dimensions of the texture features (period):

- Direct laser writing (DLW) is applied when the texture is of 10's to 100's of micrometers in the scale. The most popular texture is dimples craters ablated by single or multiple laser pulses used for lubrication and friction reduction [111], [112].
- Direct laser interference patterning (DLIP) is beneficial when a periodical texture is needed with a period from tens of micrometers down to hundreds of nanometers [73], [76].
- Laser-induced periodic surface structures (LIPSS), often termed *ripples*, are self-organized periodical structures occurring on the surface of any material under laser irradiation with linearly polarized laser radiation [113].
- Hierarchical structures are fabricated by combining a few approaches above and usually provide for the long-term functionality of laser-treated surfaces [80]–[82].

LIPSS have gained an increasing attraction as these periodic structures with a period below the wavelength of laser radiation could be generated in a simple single-step process, allowing a surface nanostructuring for tailoring optical, mechanical, and chemical surface properties [113]-[115]. Additional mechanical or chemical post-processing steps are usually not required by utilizing USP lasers for LIPSS formation. They show excellent tribological performance (friction and wear) on steel and titanium alloys [116]. Laser-induced periodic surface structures could be produced on cold work tool steel by irradiation with a low number of picosecond laser pulses [117], meaning that this kind of nanotexturing has huge potential for industrial upscaling to large areas. Highly regular LIPSS have been fabricated on surfaces of molybdenum, steel alloy and titanium at high processing speed on large areas adjusting the laser process parameters to the material properties [118]. An additional enhancement in LIPSS

formation is achieved by utilizing a burst of laser pulses with intra-burst delays on the picosecond timescale [119].

More complex surface textures could be fabricated by combining DLIP with the generation of LIPSS in a one-step process [120]. The technique provides multi-scale periodic patterns with two-dimensional symmetry. In the case of stainless steel, it is possible to create the line- or pillar-like surface textures by rotating the polarization of the laser beam. For sapphire, the interference-like texture and LIPSS were realized simultaneously. The combined spatial and temporal shaping of the pulse in DLIP increases the level of control over the texture pattern [77].

The practical application of laser-generated micro- and nanostructures that control hydrophobicity of surfaces is preventing ice formation. The problem is relevant in aerospace, energy supply, telecommunication, environment protection [121], [122]. The laser texturing strategies, including direct laser writing (DLW) to laser-induced periodic surface structuring (LIPSS) and direct laser interference patterning (DLIP), provide excellent results on metals and polymers, providing a long-living effect.

C. Processing of Transparent Materials

Transparent materials, like glass, fused silica, sapphire, are particular media where the use of ultra-short pulse lasers is beneficial due to nonlinear interaction with the material, leading to enhanced highly-localized energy deposition. That is an extremely broad area of material processing methods, and very few examples will be reviewed here.

Glass cleaving or separation is a wide-spread application. The use of USP lasers was initiated by the P. Herman group, which exploited the filamentation of femtosecond pulses with energy above the critical one [123]. The ultrafast laser burst trains provide clear benefits in generating filamentation tracks to guide the cleaving of glass substrates. The laser filament cutting of soda-lime silicate glass was validated using a picosecond pulsed Nd:YAG laser with varying burst energies [124]. The cleaving cracks were guided by the filament only if the network of microcracks is sufficiently developed. Bursts of picosecond laser pulses without filamentation initiated the evolution of directional cracks growing bottom-up in sapphire and facilitated a high-speed wafer dicing [125].

Bessel-type beams with elongated focus are proved to be beneficial in cleaving-based separation transparent materials. Bessel beam has a high-intensity hot spot with subwavelength diameter propagating over the distance exceeding 100's of micrometers. The focal line scales up the pulse energy of a laser, and stealth dicing of thick glass up to 1 cm is feasible in a single pass [126]. Directional crack formation for confined cleavage is easy controlled by deformation of the axial symmetry of the Bessel beam, which is crucial in the case of glass cutting applications [127]. Ultrafast laser cutting of a glass substrate at an oblique angle was demonstrated using a phase-corrected Bessel beam [128]. Three passes at different incident angles were combined to create a chamfer-shaped damage tract in the glass substrate. The controlled and uniform energy deposition is a critical factor for high-aspect and high-quality intra-volume modifications of

glass [129]. Upscaling of the process is feasible with high power USP lasers. An ultrafast thin-disk multi-pass laser providing up to 1.9 kW of average output power and pulse energies in the 10 mJ range at 1 ps of pulse duration was applied for glass cleaving [130]. A 3.8-mm-thick boro-float glass was separated at a speed of 1200 mm/s utilizing 300 W of input power. A single-pass modification of 12 mm-thick soda-lime glass and subsequent separation was achieved by using a 4-pulse burst with total burst energy of 1.5 mJ at a pulse duration of 3 ps [131].

The quality of laser cutting of glass is measured not only by surface roughness but mainly by the edge strength to bending. Detailed comparison of different glass cutting methods, including laser-based, is performed in [132], taking into account surface quality, side-wall roughness, residual stresses and flexural strength. Laser technologies are progressing permanently. At the optimum condition, the femtosecond-laser Bessel beam provides the front-side and back-side edge strength of 370 MPa (67% of the strength of the polished glass) and 400 MPa (72%), respectively [133].

GHz bursts of USP laser pulses provide enhanced ablation efficiency of fused silica [134]. The maximum ablation efficiency at the optimum fluence was as high as 3.05 mm³/min/W for 10-pulse bursts, which is 7.4 times higher than the single-pulse regime's efficiency (0.41 mm³/min/W). By adjusting processing regimes, USP lasers could be successfully applied in direct freeform optics fabrication.

V. CONCLUSION

Significant progress in the reliable generation, amplification and shaping of ultra-short laser pulses was achieved during recent years. Starting from the evolution of the ultra-short-pulse lasers utilized in the laser material process, the review covered application areas, which currently are emerging:

- The high-efficient ablation, scaling up the process throughput to be competitive with alternative material processing technologies;
- Processing of transparent materials, especially glass separation.

There are many more application fields not detailed here, where USP lasers are beneficial or provide unique, otherwise not achievable opportunities. The intra-volume modifications of transparent materials, which depending on excitation degree, lead to:

- Waveguide or grating inscription;
- Nanograting formation, which facilitates selective chemical etching (FLICE) of complex "ship in a bottle" structures for fluidics and micromechanics;
- Q-plates, 5D memory, etc.

New technologies and methods appear, exploring unlimited possibilities of coherent light pulses with extremely short duration.

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