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Ultra-Short-Pulse Lasers—Materials—Applications †

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Abstract: We overview recent developments of 3D$^{\pm}$ (additive/subtractive) manufacturing/printing from the point of view of laser development, beam delivery tools, applications, and materials. The average power of ultra-short-pulsed lasers has followed a Moore's scaling trajectory, doubling every two years, for the past 20 years. This requires fast beam scanning solutions and beam delivery control for larger-area applications. New material synthesis with high spatial resolution is provided at the high intensity TW/cm^2-PW/cm^2 exposure site. Net-shape manufacturing with a reduced number of post-processing steps is a practical trait of 3D$^{\pm}$ printing. With computer numerical control (CNC) optimised using artificial intelligence (AI), the future of 3D$^{\pm}$ manufacturing is discussed.

Keywords: 3D printing; ablation; light–matter interaction; femtosecond lasers; nanoscale

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1. Laser Source and Beam Delivery

1.1. Ultra-Short-Pulse Laser Evolution

Laser, as a non-contact energy delivery tool, has a unique capability harnessed for fundamental research in the inertial confinement fusion (ICF), which recently became a step closer by reaching the burning plasma condition [1]. A laser intensity increase over the years after its invention in 1960 is a constant trend important for the basic science of light–matter/plasma interactions, as outlined in a roadmap review [2]. Matter at extreme conditions at pressures above 1 megabar (10^{11} Pa) is currently one of the most active fields of research [3].

Since the year 2000, the average laser power of ultra-short (sub-1 ps) pulsed lasers has increased as $Power = 2^{N/2}$, with N being the number of years from the beginning of the trend, which parallels Moore's law for the number of transistors in an integrated circuit. This conclusion is achieved following the evolution of ultra-short-pulsed laser amplitude produced over the last 20 years, presented recently [4]. Initially based on the chirped pulse amplification (CPA), which was awarded the Nobel prize in 2018, more recent approaches exploit different cavity geometries as well as amplification via the divided pulse and coherent beam combination. These strategies further increase the extracted power from solid-state and fibre laser systems and make them more compact. Ultra-short lasers with powers in the sub-1 kW range, ~1 mJ pulse energies and at the repetition rates up to ~1 MHz have become available.

New modes of laser operation bring the capability of combining ultra-short pulses into MHz–GHz bursts with a controlled number of pulses per burst [5]. It was shown that this burst mode of operation delivers ablation rates for metal and dental tissue on the order

of 3 mm^3/min. This is the rate that reaches that of current Electrical Discharge Machining/Grinding (EDM/G) computer numerical control (CNC) tools. This parity between material removal rate by discharge spark and laser beam was achieved in 2016. The burst mode advantage is in the possibility to fine tune material removal to the most efficient fluence [J/cm^2] [6], which is empirically determined to be $e^2 = 7.4$ times larger than the ablation threshold for the given material [7]. Fine tuning the optimum ablation rate is achieved by changing the pulse number per irradiation spot, using beam scanning [8], and control over the number of pulses per burst. For comparison of different fabrication conditions, the volume [mm^3] ablated per 1 W average power per time 1 min, V_a ~mm^3/W/min ~mm^3/(W.s) ~mm^3/J, is used. This is the ablated volume-per-energy delivered by the laser for subtractive machining ($3D^{(-)}$ printing). Interestingly, we show here that the volumetric energy density *Energy*/*Volume* ~J/mm^3 is the right measure for the additive mode of $3D^{(+)}$ printing by ultra-short laser pulses [9]. It is not surprising that accounting for the energy deposition in the volume of light–matter interaction is the essential measure for the both additive and subtractive $3D^{(+)}$ and $3D^{(-)}$ modes of 3D fabrication.

1.2. Use of High-Average-Power Laser Beam

High-average-power sub-kW laser systems are targeting industrial applications. With the exponential $2^{N/2}$ increase in laser power indicated above, the most efficient use of this photon budget is required. To handle high laser power, new beam delivery systems are developed for the distribution of energy in a very well-controlled and precise way over the workpiece. Photonic crystal fibres (holy-fibres), flexible delivery units and polygon scanners with beam travel rates up to 1 km/s are readily available; interestingly, polygon scanners now used for the fastest beam delivery became available from mid-1980 and are on a similar growth trajectory to fs lasers. Galvano and polygon scanners further contribute to the compactness, versatility and safety of high-power handling. It is noteworthy that the scanning of the laser beam in cash-counter machines is an example of an application where speed and safety are delivered simultaneously. This is especially important for open-space and field-deployable applications, e.g., surface texturing by ablation ripples for the creation of hydrophobic, anti-icing and biocidal surfaces [10]. These applications are particularly suitable for fast beam scanning techniques. One of the most demanding applications for surface treatment is in the solar cells industry. Anti-reflection coatings and packaging for 20+ years continuous performance in open air have to be delivered. With the promise of increasing the efficiency of Si solar cells from the current 18% (for mass-produced cells) to one closer to the theoretical Shockley–Queisser limit of ~31%, the use of photonic crystal patterns on Si surfaces is an invitation to use fast laser scanning for laser texturing [11]. Scanning of large (cm-scale) areas without stitching errors and maintaining sub-wavelength precision of laser patterning by combined sample and beam scan was recently introduced for 3D polymerisation [12]. This approach is inherently scalable to larger (meter-scale) patterning in atmospheric (room) conditions, required for patterning surfaces for injection moulding die surfaces, texturing steel and fibre composites for anti-frosting and water repelling properties in the aviation industry, and potentially for solar cells in the future.

2. Materials

Materials are a major and critical part for the $3D^{\pm}$ manufacturing ecosystem (Figure 1). New polymerisable mixtures of colloidal particles and standard photo-polymerisable resists/resins can be tailored for the required material composition. Calcination of the polymerised composites can be transferred into a glass, polycrystalline or ceramic state with feature sizes down to the nanoscale [13]. Cutting and drilling of dielectrics, e.g., dicing of sapphire substrates in the light emitting diode (LED) industry, and metal/composite processing with high precision and minimal heat-affected zone (HAZ) for complex 3D geometries can be carried out most efficiently with ultra-short laser pulses [14]. This versatility in terms of material processing stems from well-controlled energy delivery in space and time. Even small energy pulses have high intensities—TW/cm^2 and above—and

can turn non-absorbing dielectrics into ionised plasma with strong energy deposition. Internal modification of the interior volume of dielectrics becomes feasible with these energies. It was demonstrated that high-pressure and high-temperature phases of materials can be created and retained down to room ambience due to ultra-fast thermal quenching of a small modified volume [15,16]. Internally confined micro-explosions occurring in the high-Young-modulus dielectrics create conditions similar to the centre of the Earth—hence, warm dense matter (WDM). The micro-explosion hydrodynamics follows the established and tested macroscopic versions [17]. New and metastable phases of materials, e.g., amorphous sapphire, can be produced by tightly focused fs laser pulses [18].

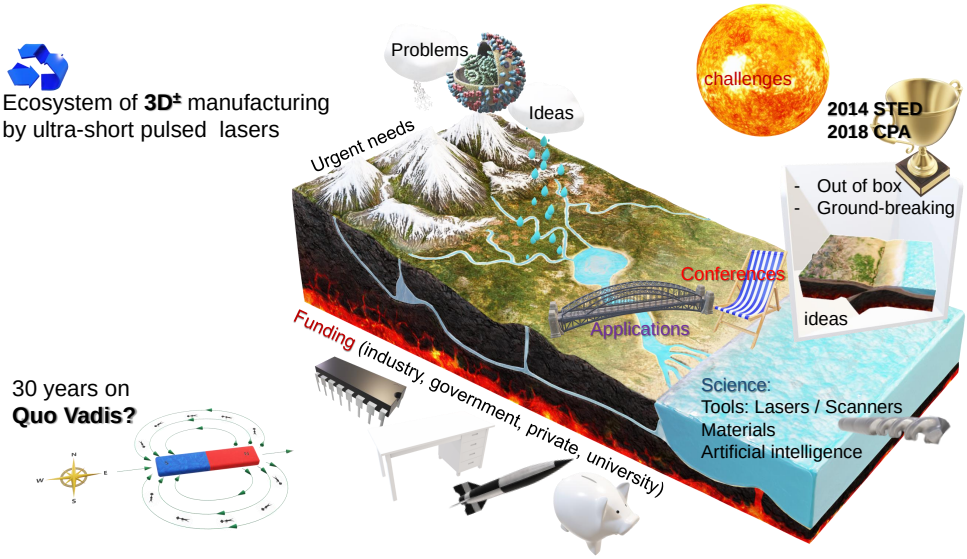

Figure 1. Ecosystem of $3D^{\pm}$ manufacturing based on development of lasers, beam/stage scanners, computer numerical control (CNC), artificial intelligence (AI). Increasing field of applications in material processing and creation of new materials is developing via different funding sources.

The mass production of colloidal nanoparticles of different materials in water with fs laser pulses scanned at speeds exceeding that of bubble formation is already an industrial process. The benefits of such nanoparticles are that surfaces are free from surfactants used in chemical synthesis. The size distribution of these colloids can be controlled via interaction with simultaneously generated coherent white light continuum (WLC) [19].

A large impact on the development of material processing by ultra-short laser pulses was driven by the quest for higher resolution—ultimately, super-resolution—which can deliver the fabrication of 3D objects with sub-diffraction λ/NA and sub-wavelength resolution; NA is the numerical aperture of the optics used, and λ is the wavelength. The method of stimulated emission depletion (STED) microscopy, demonstrated in 2000 and awarded the Nobel prize in 2014, influenced the community of fs laser users who widely relied on table-top microscopes used for the polymerisation of nano-micro-structures and optical memory. Due to the threshold effect of material modification, tens-of-nm resolution in 3D can be achieved by direct fs laser write via the fine tuning of the pulse energy. This is even without critical point drying (CPD) equipment, which is typically used to avoid deformations made by surface tension during the wet development stage; a 30 nm 3D feature size was obtained using the threshold effect in common SU8 [20].

3. Applications

Beyond material processing, ultra-short laser pulses are used in an ever-increasing range of applications, especially due to the available high power and dramatic reductions in size. Ultra-short laser pulses in the vis-IR spectral range have potential for data communication, especially in non-scattering ambience, e.g., for space applications due to

high frequency—hence, large bandwidth is required for fast data communications. It is a recognisable trend in wireless and mobile communications.

Direct energy deposition applications already range from defence to $3D^{+}$ printing (e.g., powder sintering). In the practical, high-fluence/-intensity application of laser cutting, the use of linearly shaped focal regions, e.g., Gauss–Bessel beams, is proving to be a viable solution [21,22].

In multi-dimensional optical memory, the usual 3D positioning of memory bits for laser writing and readout by luminescence or scattering [23,24] is augmented by a polarisation degree of freedom due to nano-gratings, which form two extra dimensions via form birefringence. Fs-inscribed optical memory bits withstand 1100 °C temperatures [25]. Optical memory is of significant interest due to its thermal stability and durability.

Coming full circle, for high-spatial-resolution studies with single fs laser pulses and interference patterns [26–29], the most recent development of high-precision direct write shows the possibility of fabricating nanoscale grooves down to 20 nm width on a solid-state dielectric film (equivalent of a resist) [30]. Precise energy control by the orientation of linear polarisation allows the patterning of single nanoscale features: bumps, voids and grooves [31,32].

For the commercial viability of any technical solution, it is necessary for it to deliver a bridging solution in product manufacturing that is unique: better before cheaper. Based on the commercial success of a particular implementation, other areas as well as more fundamental research are funded (Figure 1). It is increasingly difficult to make improvements to production line processes as a new project due to complications of a fast-moving industry cycle (<1 year), in contrast to academic research, which is a multi-year endeavour, e.g., can be measured in duration of PhD projects (∼3–4 years). Due to this complexity and lengthy project review (∼0.5 years), the entry point between academia and industry is most efficient for small-scale proof of the principle applications. Rapid prototyping, which is the key advantage of $3D^{\pm}$ printing by ultra-short laser pulses, is the most promising pathway for industry–academia engagement. The trend for using artificial intelligence (AI) in the CNC control of processes is rapidly evolving. Recently, predictions of the optical properties of complex 3D multilayered structures of different materials for specific spectral functions were AI generated with convincing fidelity [33].

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