Opinion Vol. 14, No. 10 / 1 Oct 2024 / Optical Materials Express 2370
Optical Materials EXPRESS

Holographic multi-photon 3D laser nanoprinting – at the speed of light: opinion

PAUL SOMERS,[1] SEBASTIAN KOCH,[2] PASCAL KIEFER,[2] MARYNA L. MERETSKA,[1] AND MARTIN WEGENER[1,2,*]

[1] *Institute of Nanotechnology (INT), Karlsruhe Institute of Technology (KIT), Karlsruhe 76131, Germany*
[2] *Institute of Applied Physics (APH), Karlsruhe Institute of Technology (KIT), Karlsruhe 76131, Germany*
[*] *martin.wegener@kit.edu*

Abstract: In this opinion article, we discuss the possibility of printing three-dimensional macroscopic architectures with nanometer feature size by irradiating a light-sensitive ink with a single, spatiotemporally shaped, short laser pulse. We argue that the peak print rate of this approach may reach 10^{20}-10^{21} voxels s^{-1}, surpassing the present state-of-the-art of about 10^8 voxels s^{-1} by a very large margin.

Three-dimensional (3D) printing is a rapidly advancing field, in which light-based approaches play a leading role [1]. A spot of light can easily be positioned inside the volume of a transparent ink to induce a chemical reaction, *e.g.*, leading to the solidification of a liquid monomer into a polymer 'voxel', a 3D volume element in analogy to the 2D pixel. After scanning the spot of light along a desired path, the remaining insufficiently crosslinked material can be washed out by a solvent, leaving behind the final 3D structure. One must be aware though that the accumulation of locally deposited dose, *i.e.*, the locally created density of chemical bonds, generally leads to an undesirable 'proximity effect' which limits the achievable printing resolution. Nonlinear processes such as two-photon absorption [2] or (1 + 1)-photon absorption [3–6], *e.g.* two-step absorption [4], can suppress this effect and are widely used. Furthermore, one can form a large number of small spots of light, which can be seen as a large assembly of 'local nozzles of light' in 3D. Various approaches have recently used this path to parallelize the otherwise purely sequential printing process. For example, by using a light-sheet approach and 33,000 independent laser spots in one plane, print rates around 10^7 voxels s^{-1} were achieved [7]. Using spatiotemporal focusing led to print rates above 10^6 voxels s^{-1} [8]. Using an array of 7×7 laser foci and focus scan speeds around 1 m s^{-1} enabled a print rate of about 10^8 voxels s^{-1} [9]. These and many other results are summarized in Fig. 1 [10].

This raises the fundamental question: What is the speed limit for 3D printing with light? One can imagine to expose all voxels of a target structure in a single short laser pulse. Spatial, or even spatiotemporal, shaping of the light field to achieve this is possible – although challenging – by creating a 3D hologram via suitable 'phase masks' (see below). However, we must consider that to polymerize a voxel by multi-photon absorption, a certain incident optical energy E is required. This energy depends on the ink (commonly a light-sensitive photoresist) used for the printing. Future progress in photochemistry can potentially offer more sensitive inks [11–13], but the value for E will remain finite. Consider the currently most sensitive photoinitiators and resists [14] based on two-photon absorption when printing with order 100 fs long laser pulses from a laser oscillator at a repetition rate of around 100 MHz and focus scan speeds of 1 m s^{-1}: It takes about 50 laser pulses with an individual pulse energy of about 0.2 nJ to polymerize one voxel [9]. For $7 \times 7 = 49$ foci in parallel, this corresponds to an average laser power of about 1 W incident on the entrance pupil of a high-numerical-aperture immersion microscope objective lens.

#538751 https://doi.org/10.1364/OME.538751
Journal © 2024 Received 6 Aug 2024; revised 28 Aug 2024; accepted 28 Aug 2024; published 17 Sep 2024

Optical Materials EXPRESS

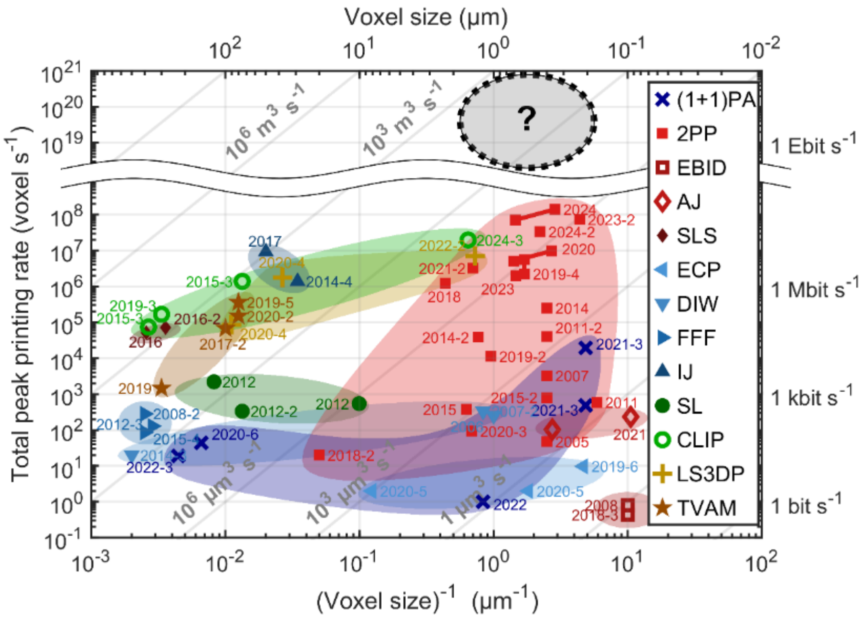

Fig. 1. Peak printing rate of various 3D printing technologies in relation to their minimum voxel size (taken as an average of the lateral and axial voxel dimensions) [10]. The region of printing speed potentially accessible via 3D multi-photon holographic laser printing is indicated in gray. Data sources for the data points are as follows: 2005 [15], 2006 [16], 2007 [17], 2007-2 [18], 2008 [19], 2008-2 [20], 2011 [21], 2011-2 [22], 2012 [23–25], 2012-2 [26], 2012-3 [27], 2014 [28], 2014-2 [29], 2014-3 [30], 2014-4 [31], 2015 [32], 2015-2 [33], 2015-3 [34], 2015-4 [35], 2016 [36], 2016-2 [37], 2017 [38], 2017-2 [39], 2018 [40], 2018-2 [41], 2018-3 [42], 2019 [43], 2019-2 [44], 2019-3 [45], 2019-4 [46], 2019-5 [47], 2019-6 [48], 2020 [10], 2020-2 [49], 2020-3 [50], 2020-4 [51], 2020-5 [52], 2020-6 [53], 2021 [54], 2021-2 [8], 2021-3 [4], 2022 [55], 2022-2 [7], 2022-3 [5], 2023 [56], 2023-2 [57], 2024 [9], 2024-2 [58], 2024-3 [59]. (1 + 1)PA, (1 + 1)-photon absorption; 2PP, two-photon polymerization; EBID, electron beam induced deposition; AJ, aerosol jet 3D nanoprinting; SLS, selective laser sintering; ECP, electrochemical printing; DIW, direct ink writing; FFF, fused filament fabrication; IJ, inkjet; SL, stereolithography; CLIP, continuous liquid interface printing; LS3DP, light-sheet 3D printing; TVAM, tomographic volumetric additive manufacturing. Figure adapted with permission from Ref. [10], CC BY 4.0.

Such powers are not far from the damage threshold of the lens, meaning that orders-of-magnitude print-speed increases cannot be expected solely along the path of delivering more laser power.

We now conceptualize fully parallelized holographic 3D printing with a single shaped laser pulse: The pulse enters the ink along the *z*-direction through a glass interface parallel to the *xy*-plane. In the wavelength-thick layer next to the substrate, the light field may, for example, be composed of many individual light spots that expose many different voxels in parallel in that single plane. We now take advantage of the fact that multi-photon absorption by the ink consumes only a tiny fraction of the pulse energy, such that nearly all of the light is transmitted through the first layer. In the above multi-focus approach, all of this transmitted light was essentially wasted because the foci of light (intentionally) diverged so much, that multi-photon absorption above and below the focal plane became extremely weak. In that multi-focus approach, a next (*e.g.* wavelength-thick) layer would be exposed by scanning the focal plane along the *z*-direction, with typical velocities not exceeding $v = 10^{-3}$ m s^{-1}. In the holographic approach, light propagates along the *z*-direction and energy is redistributed at the speed of light, about

Optical Materials EXPRESS

$c = \frac{2.998 \times 10^8 \text{ m s}^{-1}}{n} \approx 2 \times 10^8$ m s^{-1} in a medium with an example refractive index of $n = 1.5$, and hence eleven orders of magnitude faster – 3D printing at the speed of light.

The holographic shaping of the laser pulse would lead to different exposure patterns in the consecutive layers. How long would it take to expose an object that is, *e.g.*, 200 µm tall along the z-direction? The exposure time for using a single short laser pulse would be 200 µm $/c \approx 1 \times 10^{-12}$ s = 1 ps, which is 10 times longer than the 100-fs laser pulse duration in our discussion above. The print process can then be viewed as a thin layer of light scanning along the z-direction at the speed of light (Fig. 2). The polymerization induced by this light exposure would take place on a much longer time scale of ∼0.1 – 1 ms [60], determined by the speed of oxygen diffusion. This time is merely a delay and does not influence the print rate.

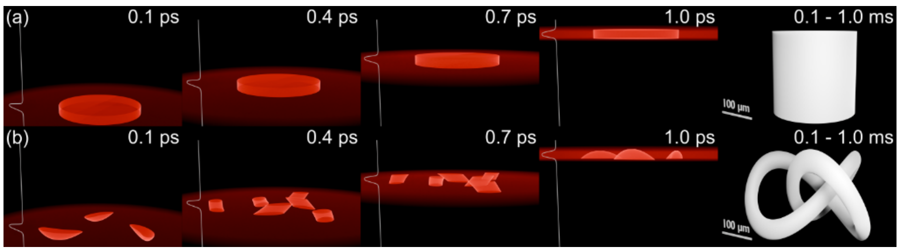

Fig. 2. Schematic of single-pulse 3D holographic printing. An artistic illustration of a single 100 fs laser pulse propagating through a photosensitive ink forming a light field intensity profile corresponding to (a) a solid cylinder and (b) a more complex 3D shape of a trefoil knot. The first four panels capture the light field at separate instances of time during the pulse propagation. The final panel depicts the resulting polymerized 3D object after a delay of order 0.1–1 ms.

Do we realistically have sufficient laser pulse energy to expose a large fraction of a typical high-numerical-aperture microscope lens' field of view in one shot? The two-photon polymerization dose is proportional to the square of the pulse energy, meaning that to go from using 50 pulses (see above) to a single pulse, we would need about 0.2 nJ $\times \sqrt{50} \approx 1.4$ nJ incident optical energy to polymerize one voxel with a diameter of about 350 nm. This is a realistic size for multi-photon printing. Suppose we target a print-field diameter of 350 µm. This means that the print-field area is $A = \left(\frac{350\ \mu\text{m}}{350\ \text{nm}}\right)^2 \approx 1 \times 10^6$ times larger than the voxel cross-section in the xy-plane. Then we need 1.4 nJ $\times \left(\frac{350\ \mu\text{m}}{350\ \text{nm}}\right)^2 \approx 1.4$ mJ incident optical energy in one laser pulse. At a repetition rate of 1 kHz, this number corresponds to an average laser power of 1.4 W. These parameters – 1.4 W average laser power, 1 kHz pulse repetition rate, 100 fs pulse duration, and 800 nm center wavelength – are readily accessible by modern regeneratively amplified femtosecond lasers [61,62].

What is the resulting peak print rate in units of voxels s^{-1}? Following the above estimate, we have $\left(\frac{350\ \mu\text{m}}{350\ \text{nm}}\right)^2 \approx 1 \times 10^6$ voxels in one print plane. For the voxel height, we assume one wavelength – which appears fair for conditions of high-numerical-aperture focusing. At 800 nm free-space wavelength and an ink refractive index of $n = 1.5$, this leads to 533 nm voxel height. For a 200 µm tall printed structure (see above), we can print 10^6 voxels in each layer by reusing the transmitted pulse energy from previous layers, corresponding to a total of 1×10^6 voxels $\times \frac{200\ \mu\text{m}}{533\ \text{nm}} \approx 3.8 \times 10^8$ voxels. These voxels are printed in the pulse propagation time of 1 ps, leading to a peak print rate of 3.8×10^8 voxels / 1 ps $\approx 3.8 \times 10^{20}$ voxels s^{-1}. So far, the thought experiment is naïve and conservative, as we have essentially considered printing an elongated cylinder with a 100% filling fraction of material, such as by letting a collimated laser beam pass through the ink (Fig. 2(a)). For lower filling fractions corresponding

Optical Materials EXPRESS

to more interesting 3D objects to be printed (Fig. 2(b)), the required laser pulse energy and, hence, the required average laser power would decrease in our favor. At fixed power, the addressable print volume would increase in our favor. This also means that the print rate becomes structure-dependent. With shorter laser pulses, sub-100% filling fractions, slightly more sensitive inks, or, *e.g.*, three-photon absorption instead of two-photon absorption, our estimation brings us to tantalizing peak print rates exceeding 10^{21} voxels s^{-1}. One might argue, however, that the next print event only takes place with the following laser pulse, which arrives $\frac{1}{1\,\text{kHz}} = 1$ ms later. This leads to an *average* print rate lower by a factor 1 ms / 1 ps $\approx 1 \times 10^9$, yielding an *average* print rate of 3.8×10^{11} voxels s^{-1}. That is still more than three orders of magnitude larger than the current state-of-the-art *peak* printing rate of 10^8voxels s^{-1} (see above) or about four orders of magnitude larger than the current state-of-the-art *average* printing rate.

As mentioned above, holography [63] allows to create the required shaped light fields by imposing a tailored phase front onto an incident light field. Diffractive optical elements can and have been used for spatial phase-front shaping (see, *e.g.*, [9]). Spatial liquid-crystal light modulators [64] or digital mirror devices [65] can be used for dynamic phase-front shaping, but their precision, bandwidth, and number of pixels is limited. Advanced spatiotemporal light shaping by optical metasurfaces has recently been demonstrated [66–71]. However, even designing such 'synthetic optical holograms' is a non-trivial task as it requires solving an inverse problem. Traditionally, iterative algorithms such as the Gerchberg-Saxton algorithm for reconstructing phase information of a target intensity profile are used [72]. Alternatively, one could calculate the light field resulting from an optical system and minimize a carefully chosen loss function [73]. In the end, assuming the use of 32-bit floating point numbers, one must be able to handle a target light field composed of at least $10^8 \times 4$ bytes ≈ 400 MB, which doubles to 800 MB when one considers that complex numbers are needed. This is a computationally demanding task. Of course, for a single static 3D hologram, it only needs to be solved once. To determine the solution for many holograms, which is required in the dynamic case, it may be helpful to look towards machine-learning methods [74]. No matter what, it is clear that extensive computational resources would be required, though within the realm of what is available.

Finally, we ask whether steps in the direction of holographic 3D printing with a single pulse have been taken? Perhaps one of the earliest implementations is nanosecond-pulsed laser interference lithography [75], a rather simple implementation of holography. It uses one-photon absorption and is limited to periodic structures, yet it already approaches printing 3D structures at much larger print rates than other approaches summarized in Fig. 1 [1]. However, implementations and print rates have been more modest for the more interesting case of printing with arbitrarily shaped 3D light fields. A popular implementation of holography is to split a laser beam into many foci [9,10,22,32,33,50,76–78] with the foci usually constrained to one 2D plane, with exceptions [44,56,79,80]. Other works have used holography to generate more complex light fields also confined to a 2D plane [81–84]. This has recently been extended to multiple exposures at different depths or angles to form a 3D structure in tomographic volumetric additive manufacturing processes [85,86]. Only a few works have thus far tackled the challenge of 3D printing directly with a single tailored arbitrary 3D light intensity profile [87,88], but they were restricted to small print volumes and long exposure times, thus to rather low print rates.

We conclude then, that through the combination of mJ pulse energies from commercially available amplified femtosecond laser systems and the rapidly developing field of optical metasurfaces for light-field shaping, we foresee 3D holographic multi-photon nanoprinting using a single laser pulse to have the potential for many orders of magnitude increase in peak print rate compared to the current state-of-the-art.

Funding. Deutsche Forschungsgemeinschaft (Emmy Noether 513531433, Excellence Cluster EXC-2082/1-390761711); Carl-Zeiss-Stiftung (Carl-Zeiss-Foundation-Focus@HEiKA); Helmholtz Association Materials Systems Engineering; Karlsruhe School of Optics and Photonics; Max Planck School of Photonics; Karlsruhe Institute of Technology (Young Investigator Network (YIN)); Hector Fellow Academy.

Optical Materials EXPRESS

Disclosures. The authors declare no conflicts of interest.

Data Availability. All the data generated in this paper are explicitly provided.

References

1. P. Somers, A. Münchinger, S. Maruo, *et al.*, "The physics of 3D printing with light," Nat. Rev. Phys. **6**(2), 99–113 (2023).
2. S. Maruo, O. Nakamura, and S. Kawata, "Three-dimensional microfabrication with two-photon-absorbed photopolymerization," Opt. Lett. **22**(2), 132 (1997).
3. J. Méndez-Ramos, J. C. Ruiz-Morales, P. Acosta-Mora, *et al.*, "Infrared-light induced curing of photosensitive resins through photon up-conversion for novel cost-effective luminescent 3D-printing technology," J. Mater. Chem. C **4**(4), 801–806 (2016).
4. V. Hahn, T. Messer, N. M. Bojanowski, *et al.*, "Two-step absorption instead of two-photon absorption in 3D nanoprinting," Nat. Photon. **15**(12), 932–938 (2021).
5. S. N. Sanders, T. H. Schloemer, M. K. Gangishetty, *et al.*, "Triplet fusion upconversion nanocapsules for volumetric 3D printing," Nature **604**(7906), 474–478 (2022).
6. V. Hahn, N. M. Bojanowski, P. Rietz, *et al.*, "Challenges and Opportunities in 3D Laser Printing Based on (1 + 1)-Photon Absorption," ACS Photonics **10**(1), 24–33 (2023).
7. V. Hahn, P. Rietz, F. Hermann, *et al.*, "Light-sheet 3D microprinting via two-colour two-step absorption," Nat. Photon. **16**(11), 784–791 (2022).
8. P. Somers, Z. Liang, J. E. Johnson, *et al.*, "Rapid, continuous projection multi-photon 3D printing enabled by spatiotemporal focusing of femtosecond pulses," Light: Sci. Appl. **10**(1), 199 (2021).
9. P. Kiefer, V. Hahn, S. Kalt, *et al.*, "A multi-photon (7 × 7)-focus 3D laser printer based on a 3D-printed diffractive optical element and a 3D-printed multi-lens array," Light: Advanced Manufacturing **4**(1), 1 (2024).
10. V. Hahn, P. Kiefer, T. Frenzel, *et al.*, "Rapid Assembly of Small Materials Building Blocks (Voxels) into Large Functional 3D Metamaterials," Adv. Funct. Mater. **30**(26), 1907795 (2020).
11. T. Wloka, M. Gottschaldt, and U. S. Schubert, "From Light to Structure: Photo Initiators for Radical Two-Photon Polymerization," Chem. - Eur. J. **28**(32), e202104191 (2022).
12. T. Liu, P. Tao, X. Wang, *et al.*, "Ultrahigh-printing-speed photoresists for additive manufacturing," Nat. Nanotechnol. **19**(1), 51–57 (2024).
13. S. C. Gauci, A. Vranic, E. Blasco, *et al.*, "Photochemically Activated 3D Printing Inks: Current Status, Challenges, and Opportunities," Adv. Mater. **36**(3), 2306468 (2024).
14. P. Kiefer, V. Hahn, M. Nardi, *et al.*, "Sensitive Photoresists for Rapid Multiphoton 3D Laser Micro- and Nanoprinting," Adv Opt Mater **8**(19), 2000895 (2020).
15. J. Kato, N. Takeyasu, Y. Adachi, *et al.*, "Multiple-spot parallel processing for laser micronanofabrication," Appl. Phys. Lett. **86**(4), 044102 (2005).
16. G. M. Gratson, F. García-Santamaría, V. Lousse, *et al.*, "Direct-write assembly of three-dimensional photonic crystals: Conversion of polymer scaffolds to silicon hollow-woodpile structures," Adv. Mater. **18**(4), 461–465 (2006).
17. X. Z. Dong, Z. S. Zhao, and X. M. Duan, "Micronanofabrication of assembled three-dimensional microstructures by designable multiple beams multiphoton processing," Appl. Phys. Lett. **91**(12), 1–4 (2007).
18. E. B. Duoss, M. Twardowski, and J. A. Lewis, "Sol-Gel Inks for Direct-Write Assembly of Functional Oxides," Adv. Mater. **19**(21), 3485–3489 (2007).
19. H. Plank, C. Gspan, M. Dienstleder, *et al.*, "The influence of beam defocus on volume growth rates for electron beam induced platinum deposition," Nanotechnology **19**(48), 485302 (2008).
20. "Stratasys Fortus 360MC. (2008).," (n.d.).
21. J. Fischer and M. Wegener, "Three-dimensional direct laser writing inspired by stimulated-emission-depletion microscopy [Invited]," Opt. Mater. Express **1**(4), 614 (2011).
22. S. D. Gittard, A. Nguyen, K. Obata, *et al.*, "Fabrication of microscale medical devices by two-photon polymerization with multiple foci via a spatial light modulator," Biomed. Opt. Express **2**(11), 3167 (2011).
23. X. Zheng, J. Deotte, M. P. Alonso, *et al.*, "Design and optimization of a light-emitting diode projection micro-stereolithography three-dimensional manufacturing system," Rev. Sci. Instrum. **83**(12), 125001 (2012).
24. X. Zheng, H. Lee, T. H. Weisgraber, *et al.*, "Ultralight, ultrastiff mechanical metamaterials," Science **344**(6190), 1373–1377 (2014).
25. X. Zheng, W. Smith, J. Jackson, *et al.*, "Multiscale metallic metamaterials," Nat. Mater. **15**(10), 1100–1106 (2016).
26. "ProJet 7000 HD," Vendor Information (2012).
27. "Stratasys Mojo," (2012).
28. "Nanoscribe Photonic Professional GT," Datasheet (2014).
29. T. Bückmann, M. Thiel, M. Kadic, *et al.*, "An elasto-mechanical unfeelability cloak made of pentamode metamaterials," Nat. Commun. **5**(1), 4130–4136 (2014).
30. E. B. Duoss, T. H. Weisgraber, K. Hearon, *et al.*, "Three-dimensional printing of elastomeric, cellular architectures with negative stiffness," Adv. Funct. Mater. **24**(31), 4905–4913 (2014).
31. "Stratasys Objet 500 Connex1," 2014 (n.d.).
32. L. Yang, A. El-Tamer, U. Hinze, *et al.*, "Parallel direct laser writing of micro-optical and photonic structures using spatial light modulator," Opt Lasers Eng **70**, 26–32 (2015).

Optical Materials EXPRESS

33. W. Yan, B. P. Cumming, and M. Gu, "High-throughput fabrication of micrometer-sized compound parabolic mirror arrays by using parallel laser direct-write processing," J. Opt. **17**(7), 075803 (2015).
34. J. R. Tumbleston, D. Shirvanyants, N. Ermoshkin, *et al.*, "Continuous liquid interface production of 3D objects," Science **347**(6228), 1349–1352 (2015).
35. "Ultimaker 2 Extended," (2015).
36. "EOS P 770 with PA 2200 Top Speed 1.0," Datasheet and Vendor information (2016).
37. "EOS P 110 with PA 2200 Top Quality 1.0," Datasheet and Vendor Information (2016).
38. "HP Jet Fusion 3D 4210," Datasheet (2017).
39. M. Shusteff, A. E. M. Browar, B. E. Kelly, *et al.*, "One-step volumetric additive manufacturing of complex polymer structures," Sci. Adv. **3**(12), eaao5496 (2017).
40. B. W. Pearre, C. Michas, J.-M. Tsang, *et al.*, "Fast Micron-Scale 3D Printing with a Resonant-Scanning Two-Photon Microscope," Addit. Manuf. **30**, 100887 (2019).
41. W. Chu, Y. Tan, P. Wang, *et al.*, "Centimeter-Height 3D Printing with Femtosecond Laser Two-Photon Polymerization," Adv Mater Technol **3**(5), 1700396 (2018).
42. L. Keller and M. Huth, "Pattern generation for direct-write three-dimensional nanoscale structures via focused electron beam induced deposition," Beilstein J. Nanotechnol. **9**(1), 2581–2598 (2018).
43. B. E. Kelly, I. Bhattacharya, H. Heidari, *et al.*, "Volumetric additive manufacturing via tomographic reconstruction," Science **363**(6431), 1075–1079 (2019).
44. Q. Geng, D. Wang, P. Chen, *et al.*, "Ultrafast multi-focus 3-D nano-fabrication based on two-photon polymerization," Nat. Commun. **10**(1), 2179 (2019).
45. D. A. Walker, J. L. Hedrick, and C. A. Mirkin, "Rapid, large-volume, thermally controlled 3D printing using a mobile liquid interface," Science **366**(6463), 360–364 (2019).
46. S. K. Saha, D. Wang, V. H. Nguyen, *et al.*, "Scalable submicrometer additive manufacturing," Science **366**(6461), 105–109 (2019).
47. P. Nuñez Bernal, P. Delrot, D. Loterie, *et al.*, "Volumetric Bioprinting of Complex Living-Tissue Constructs within Seconds," Adv. Mater. **31**(42), 1904209 (2019).
48. A. Reiser, M. Lindén, P. Rohner, *et al.*, "Multi-metal electrohydrodynamic redox 3D printing at the submicron scale," Nat. Commun. **10**(1), 1853 (2019).
49. D. Loterie, P. Delrot, and C. Moser, "High-resolution tomographic volumetric additive manufacturing," Nat. Commun. **11**(1), 852 (2020).
50. C. Maibohm, O. F. Silvestre, J. Borme, *et al.*, "Multi-beam two-photon polymerization for fast large area 3D periodic structure fabrication for bioapplications," Sci Rep **10**(1), 8740 (2020).
51. M. Regehly, Y. Garmshausen, M. Reuter, *et al.*, "Xolography for linear volumetric 3D printing," Nature **588**(7839), 620–624 (2020).
52. G. Ercolano, T. Zambelli, C. van Nisselroy, *et al.*, "Multiscale Additive Manufacturing of Metal Microstructures," Adv Eng Mater **22**(2), 1900961 (2020).
53. Y. Chen, J. Zhang, X. Liu, *et al.*, "Noninvasive in vivo 3D bioprinting," Sci. Adv. **6**(23), eaba7406 (2020).
54. W. Jung, Y.-H. Jung, P. V. Pikhitsa, *et al.*, "Three-dimensional nanoprinting via charged aerosol jets," Nature **592**(7852), 54–59 (2021).
55. D. K. Limberg, J. H. Kang, and R. C. Hayward, "Triplet-Triplet Annihilation Photopolymerization for High-Resolution 3D Printing," J. Am. Chem. Soc. **144**(12), 5226–5232 (2022).
56. W. Ouyang, X. Xu, W. Lu, *et al.*, "Ultrafast 3D nanofabrication via digital holography," Nat. Commun. **14**(1), 1716 (2023).
57. B. J. Jiao, F. Chen, Y. Liu, *et al.*, "Acousto-optic Scanning Spatial-switching Multiphoton Lithography," Int. J. Extrem. Manuf. **5**(1), 035008 (2023).
58. L. Zhang, C. Wang, C. Zhang, *et al.*, "High-Throughput Two-Photon 3D Printing Enabled by Holographic Multi-Foci High-Speed Scanning," Nano Lett. **24**(8), 2671–2679 (2024).
59. J. M. Kronenfeld, L. Rother, M. A. Saccone, *et al.*, "Roll-to-roll, high-resolution 3D printing of shape-specific particles," Nature **627**(8003), 306–312 (2024).
60. J. B. Mueller, J. Fischer, F. Mayer, *et al.*, "Polymerization Kinetics in Three-Dimensional Direct Laser Writing," Adv. Mater. **26**(38), 6566–6571 (2014).
61. "Solstice Ace Ultrafast Amplifiers," https://www.spectra-physics.com/en/f/solstice-ace-ultrafast-amplifier.
62. "Coherent Astrella," https://www.coherent.com/lasers/amplifiers/astrella.
63. A. Bañas and J. Glückstad, "Light Shaping with Holography, GPC and Holo-GPC," Optical Data Processing and Storage **3**(1), 20–40 (2017).
64. "Spatial Light Modulators," https://holoeye.com/products/spatial-light-modulators/.
65. "Texas Instruments DLP Devices," https://www.mouser.com/new/texas-instruments/ti-dlp/.
66. X. Ni, A. V. Kildishev, and V. M. Shalaev, "Metasurface holograms for visible light," Nat. Commun. **4**(1), 2807 (2013).
67. G. Zheng, H. Mühlenbernd, M. Kenney, *et al.*, "Metasurface holograms reaching 80% efficiency," Nat Nanotechnol **10**(4), 308–312 (2015).
68. A. C. Overvig, S. Shrestha, S. C. Malek, *et al.*, "Dielectric metasurfaces for complete and independent control of the optical amplitude and phase," Light: Sci. Appl. **8**(1), 92 (2019).

Optical Materials EXPRESS

69. Y. Hu, X. Luo, Y. Chen, *et al.*, "3D-Integrated metasurfaces for full-colour holography," Light: Sci. Appl. **8**(1), 86 (2019).
70. N. A. Rubin, A. Zaidi, A. H. Dorrah, *et al.*, "Jones matrix holography with metasurfaces," Sci. Adv. **7**(33), 7488–7501 (2021).
71. S. W. D. Lim, J.-S. Park, D. Kazakov, *et al.*, "Point singularity array with metasurfaces," Nat. Commun. **14**(1), 3237 (2023).
72. R. W. Gerchberg and W. O. Saxton, "A Practical Algorithm for the Determination of Phase from Image and Diffraction Plane Pictures," Optik (Stuttg) **35**(2), 237–246 (1972).
73. M. M. R. Elsawy, S. Lanteri, R. Duvigneau, *et al.*, "Numerical Optimization Methods for Metasurfaces," Laser Photon Rev **14**(10), 1900445 (2020).
74. P. R. Wiecha, A. Arbouet, C. Girard, *et al.*, "Deep learning in nano-photonics: inverse design and beyond," Photonics Res **9**(5), B182 (2021).
75. M. Campbell, D. N. Sharp, M. T. Harrison, *et al.*, "Fabrication of photonic crystals for the visible spectrum by holographic lithography," Nature **404**(6773), 53–56 (2000).
76. K. Obata, J. Koch, U. Hinze, *et al.*, "Multi-focus two-photon polymerization technique based on individually controlled phase modulation," Opt. Express **18**(16), 17193 (2010).
77. B. Xu, W.-Q. Du, J.-W. Li, *et al.*, "High efficiency integration of three-dimensional functional microdevices inside a microfluidic chip by using femtosecond laser multifoci parallel microfabrication," Sci Rep **6**(1), 19989 (2016).
78. Z.-Y. Zhang, C.-C. Zhang, Y.-L. Hu, *et al.*, "Highly uniform parallel microfabrication using a large numerical aperture system," Appl. Phys. Lett. **109**(2), 021109 (2016).
79. G. Vizsnyiczai, L. Kelemen, and P. Ormos, "Holographic multi-focus 3D two-photon polymerization with real-time calculated holograms," Opt. Express **22**(20), 24217 (2014).
80. M. Manousidaki, D. G. Papazoglou, M. Farsari, *et al.*, "3D holographic light shaping for advanced multiphoton polymerization," Opt. Lett. **45**(1), 85 (2020).
81. L. Yang, J. Li, Y. Hu, *et al.*, "Projection two-photon polymerization using a spatial light modulator," Opt. Commun. **331**, 82–86 (2014).
82. L. Yang, D. Qian, C. Xin, *et al.*, "Two-photon polymerization of microstructures by a non-diffraction multifoci pattern generated from a superposed Bessel beam," Opt. Lett. **42**(4), 743 (2017).
83. N. J. Jenness, R. T. Hill, A. Hucknall, *et al.*, "A versatile diffractive maskless lithography for single-shot and serial microfabrication," Opt. Express **18**(11), 11754 (2010).
84. C. Zhang, Y. Hu, W. Du, *et al.*, "Optimized holographic femtosecond laser patterning method towards rapid integration of high-quality functional devices in microchannels," Sci Rep **6**(1), 33281 (2016).
85. C. C. Li, J. Toombs, V. Subramanian, *et al.*, "Multi-beam phase mask optimization for holographic volumetric additive manufacturing," arXiv, (2024).
86. M. I. Álvarez-Castaño, A. G. Madsen, J. Madrid-Wolff, *et al.*, "Holographic Volumetric Additive Manufacturing," arXiv, (2024).
87. D. Yang, L. Liu, Q. Gong, *et al.*, "Rapid Two-Photon Polymerization of an Arbitrary 3D Microstructure with 3D Focal Field Engineering," Macromol. Rapid Commun. **40**(8), 1900041 (2019).
88. L. Liu, D. Yang, W. Wan, *et al.*, "Fast fabrication of silver helical metamaterial with single-exposure femtosecond laser photoreduction," Nanophotonics **8**(6), 1087–1093 (2019).