Views

Anya Grushina*

Direct-write grayscale lithography

https://doi.org/10.1515/aot-2019-0024
Received February 28, 2019; accepted April 23, 2019; previously published online May 24, 2019

Abstract: Grayscale lithography is used to produce three-dimensional (3D) structures on micro- and nanoscale. During the last decade, micro-optics and other applications were actively pushing the market demand for such structures. Direct-write systems that use lasers and heated scanning probes can be used for high-precision grayscale micro- and nanolithography. They provide solutions for the most demanding applications in research and industrial manufacturing. At both the micro- and nanoscale, though, some challenges remain, mainly related to throughput. Ongoing R&D efforts and emerging new applications drive several companies to join forces in order to meet the market demands for grayscale lithography of today and in the future.

Keywords: micro-optical devices; microstructure fabrication; nanostructure fabrication; three-dimensional lithography.

1 Introduction

We routinely take pictures with our smartphones and rarely think about the progress in micro-optics that has enabled such technology. This amazing development in the fabrication of microlens arrays extends to other three-dimensional (3D) micro- and even nanostructures, too. Today, they are indispensable for numerous micro-optic and photonic applications: 3D-patterned sapphire substrates for LEDs [1], reflectors [2], blazed gratings for waveguides [3], Fresnel lenses [4, 5], holograms [6, 7], and other kinds of diffractive optical elements [8] – these are just a few examples. Besides microoptics, other technological domains also benefit from small-scale 3D elements: solar panels [4, 5], smart surfaces [8–10], IoT (Internet of things) [11], AR/VR (augmented/virtual reality) glasses, medical devices, and sensors [12]. These technologies drive the increasing demand for high-precision 3D micro- and nanostructure manufacturing, and also more intensive R&D in the field of grayscale lithography.

There are various approaches to 3D microstructure fabrication. High-precision mechanical milling and polishing can produce 3D microcomponents for optics with impressive precision [13]. However, limited throughput and resolution complicate the scalability of micromechanical manufacturing. Building up on the progress in semiconductor chip making, photolithography can be used to create 3D microstructures. Polymer resist films are exposed to varying levels of light intensity to reproduce the so-called gray levels and are further processed by well-controlled wet chemical development. High throughput in photolithography is usually achieved using photomasks as templates. They are replicated in the resist by large-area exposure to light. In the case of grayscale photolithography, very complex masks are required in order to locally modulate the light intensity. It is difficult and expensive to make such masks and to ensure that the resulting structures meet precision requirements. Even with the correct mask, such grayscale photolithography process is very hard to control [14] and is also quite limited in the shapes that it can produce [15]. Direct laser grayscale writing offers an alternative to mask-based lithography. This method has improved a lot over the last years and has proven useful for many applications, especially micro-optics.

Today, photonics and optoelectronics often require 3D nanostructures. Fabrication of such components is beyond the reach for usual photolithography. Two-photon lithography – enabled by systems from Nanoscribe, Multiphoton Optics, or Femtika – uses femtosecond lasers and can reach resolutions below 200 nm. This approach is capable of making truly 3D structures. However, challenges like resist shrinking [16] and very low throughput, still limit two-photon lithography to very small patterning volumes relevant mostly to research applications. Electron or focused ion beams can be used for even smaller 3D features, though with significant difficulties and limitations related to charging and proximity effects. A more accurate alternative approach is offered for grayscale

***Corresponding author: Anya Grushina,** Heidelberg Instruments Mikrotechnik GmbH, Heidelberg, Germany,
e-mail: grushina@swisslitho.com.
https://orcid.org/0000-0002-8749-938X

www.degruyter.com/aot

nanopatterning by thermal scanning probe lithography, which is described in more detail later in this article.

The demand for 3D micro- and nanostructures is increasing, and the number of applications using them is growing. In line with this trend, a consortium seeking a synergy in grayscale lithography development has emerged. In 2018, Heidelberg Instruments – an expert in direct laser write solutions – joined forces with SwissLitho, a pioneer in systems for thermal scanning probe lithography, and GenISys, a developer of lithographic software.

This article describes the challenges that this consortium addresses in order to meet the emerging demands from the industry and R&D for direct-write grayscale micro- and nanolithography by improving precision, throughput, and resolution.

2 Direct laser write grayscale lithography

Maskless writing appeared as an alternative to the established mask-based photolithography. Direct laser writers began substituting mask aligners and even steppers, as they could be used not only to make new masks but also to pattern the structures directly onto wafers skipping the masks entirely. This development enabled rapid and affordable prototyping, compared to mask-based photolithography. Moreover, the throughput of optical direct-write systems tremendously increased over the last years. The new generation of direct-write machines are so fast that they are used for industrial manufacturing of devices that previously could be produced with sufficient throughput only using mask-based photolithography. The progress of the direct-write approach, in general, enabled remarkable development of direct-write grayscale lithography, as some examples shown in Figure 1 illustrate. Now that the fabrication of 3D microstructures using direct laser writing is established, it calls increasingly more attention from the industry. Direct laser writing tools reflect this trend: there are solutions suitable for substrate sizes from small wafer pieces to G8 platforms (2160 × 2460 mm wafers for flat panel displays). The capability to expose areas up to 1400 × 1400 mm^2 enables the full bandwidth of applications from R&D prototyping to mass production of optical devices.

The key to grayscale lithography is to precisely control the local exposure of the resist. An example of how this

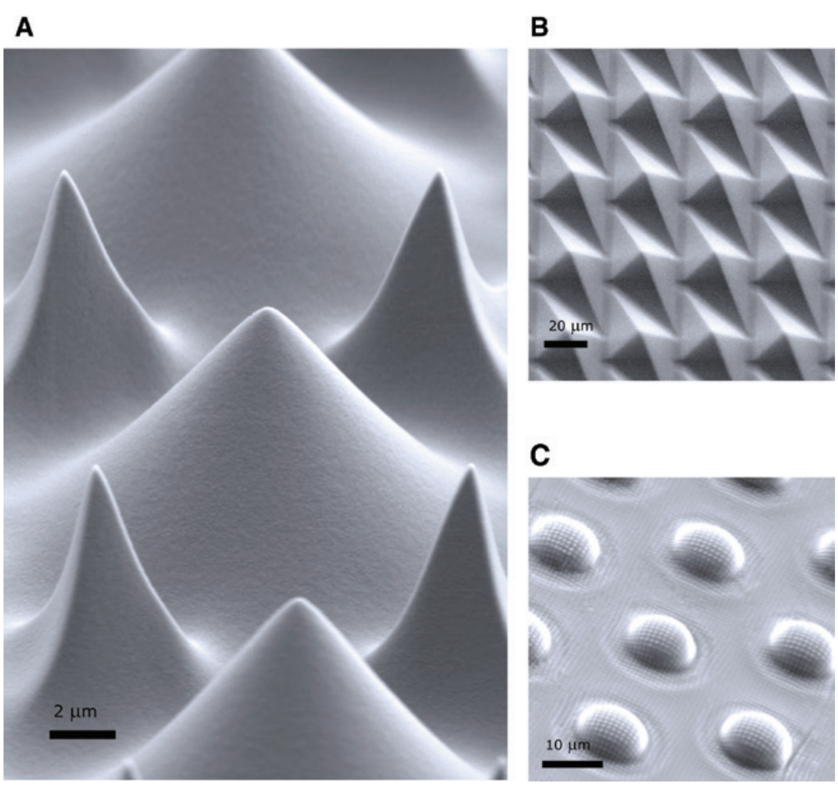

Figure 1: Examples of the grayscale lithography applications made by the DWL-series systems from Heidelberg Instruments: (A) Diffraction optical elements, IGI; (B) "Moth-eye" microlenses array, ShenZhen Nahum-Eli Optical Technology Inc, (C) Retro-reflector design, karmic.ch.

can be done is the solution implemented in DWL (direct-write laser) systems from Heidelberg Instruments (Heidelberg Instruments Mikrotechnik GmbH, Heidelberg, Germany). The optical 'heart' of these machines that enables the grayscale patterning is a spatial light modulator consisting of an acousto-optic modulator (AOM) and an acousto-optic deflector (AOD). AOMs are acousto-optical crystals connected to a piezoelectric transducer driven by the electric signal in the radiofrequency range. Resulting vibrations are transmitted into the crystal as sound waves, creating local changes in its optical density. The light passing through the crystal is diffracted, so the total light intensity can be modulated. Being fully analog, this process allows an extremely wide range of grayscale values. Having passed the AOM, the light enters the AOD. It also consists of an acousto-optical crystal and a piezoelectric transducer. Varying the frequency of the acoustic oscillations changes the direction in which the laser beam propagates. As a result, the spatial light modulator defines the exposure depth by controlling the laser light intensity and scans the substrate 'line by line', the write field being essentially a stripe. Ultimately, the resulting vertical resolution is limited by the surface roughness. All gray values are exposed in a single step, so there is no correlation between how many of them are used and the process throughput.

The lateral resolution in grayscale lithography is influenced by many factors. Instrument-wise, the main parameters affecting the resolution are the laser wavelength (set at 405 nm in DWL systems, Heidelberg Instruments Mikrotechnik GmbH, Heidelberg, Germany) and the write mode used for the exposure. The write mode is a combination of pixel size and the write lens used (one machine can feature several write modes). A typical setup for grayscale lithography produces a spot size of about 800 nm in diameter. The pixel size also determines the base throughput of the tool. Other parameters that influence the resolution are process-related: photo-resist thickness, contrast, developer, and developing time.

3 Technical challenges of grayscale lithography

The target resist profile is not so straightforward to achieve, mainly due to the complex non-linear interactions of the resist with the light. There are two main approaches to address this challenge. The first is to characterize the resist response curve and recalculate the assigned gray values (translated into the dose levels) accordingly. The second approach is to redesign the shape of the structure taking into account the resist properties and the proximity effect (features of the written pattern receive additional 'unwanted' photons from exposed features in proximity comparable with the limit of the machine's resolution). The 3D proximity effect corrections (3D PEC) are computationally very expensive. In particular, large patterns and complex geometries require specialized software to compute the corrections with sufficient accuracy and in a reasonable time. The Beamer software from GenISys (GenISys GmbH, Munich, Germany) can do 3D PEC very well, and it has an excellent reputation in the lithography community for more than 10 years. Originally designed for calculating the proximity effect corrections for e-beam lithography, this software can also model the interactions of the resist with the light. Optimized exposure pattern and grayscale values yield the desired target resist shape.

Stitching is another important challenge for direct-write lithography. The patterning is usually done pixel by pixel within a write field. When moving to the next write field, small positioning errors can occur, leading to errors in the final pattern (stitching errors). Such errors can be minimized by the ultra-precise control of the sample position with respect to the laser. In case of grayscale lithography, there is an additional challenge for stitching: at the edges of individual write fields, the potential positioning error can lead to unaccounted additional or missing exposure at the edges even when the stitching is very good. This cannot be computationally compensated; hence, small height errors at the write field edges can occur, threatening to significantly reduce the performance of a device. Such stitching errors can be avoided by the so-called 'n-over' approach: original write fields are substituted by n overlapping write fields, and the intensity is correspondingly reduced. This way, potential errors at the edges are averaged away, and at high enough n (typically $n = 4$ or higher), no stitching errors can be detected in the resulting structures anymore. An obvious downside of the 'n-over' method is the n-fold sacrifice in throughput.

Resist thickness is another limitation for the direct laser write grayscale lithography. Binary (2D) photolithography can produce high-aspect ratio microfeatures up to almost 1 mm with perfectly straight walls. At 405 nm – the laser wavelength used in DWL (Heidelberg Instruments Mikrotechnik GmbH, Heidelberg, Germany) – the maximum feature height for accurate grayscale lithography is currently around 60 µm (at a shorter wavelength the features, height is limited by 10 µm). Some applications require higher features, which now can be achieved either by pattern amplification (using reactive ion etching, for example) or by further optimization of the resists and resist process parameters.

The largest substrates relevant for industrial applications that can be patterned using direct-write grayscale lithography exceed 1 m^2. It can take several days to fully expose such a large area. To suit the industrial manufacturing processes, the direct-write grayscale photolithography needs to scale up. Possible solutions for increasing the throughput are to improve the speed of the light modulators and enable parallel exposure. Digital mirror devices (DMD) or grating light valves (GLV) are used for higher-throughput direct-write systems that are already replacing mask aligners or steppers for 2D applications. Next-generation grayscale lithography machines could rely on similar solutions. Just like in the case of mask aligners for 2D, grayscale lithography with sufficiently high throughput will remove the need for replicating techniques. As for now, nanoimprint lithography and injection molding are still required for high-throughput manufacturing of 3D microdevices.

4 Pushing the vertical resolution to a single nanometer

The miniaturization trends do not stop at the microscale. Modern circuits feature elements with a pitch of a few nanometers, and the producers of industrial equipment are pushing hard to develop reliable solutions in order to keep up. It is no surprise that 3D nanostructures also become industrially relevant for applications beyond ‘standard’ diffractive optical elements like computer-generated holograms. For example, 3D multi-mode waveguides and grating couplers for Si photonics show a significant reduction in optical losses and promise significant energy savings for data centers [17]. Nanofluidic devices with precisely engineered 3D surface patterns that can move and sort nanoobjects promise to play an important role in future diagnostic devices [18]. Optical microcavities with small mode volumes require 3D profiles defined with single-nanometer precision to maintain a high Q-factor and to be useful for quantum optics and single-photon sources [19]. For most such applications, every nanometer has a strong effect on the physical behavior of the system. While such sensitivity to the dimensions enables fine tuning of the desired properties, it is also one of the limiting factors for the wider use of such nanodevices. Until recently, 3D nanostructures could hardly be patterned with sufficient precision even in research facilities.

Electron beam lithography (EBL) is an obvious candidate for grayscale direct nanopatterning, as EBL is a well-established technique for nanofabrication in 2D. The technique has improved over several decades, and today’s high-end EBL machines, software [like BEAMER from GenISys (GenISys GmbH, Munich, Germany)], and development processes overcome most of the physical challenges of this demanding technique [20]. However, limitations of the method are strongly enhanced in the case of grayscale lithography [21]. Besides the usual issues related to charging on non-conducting substrates, proximity effects complicate the grayscale patterning with EBL tremendously, so the 3D-PEC calculation becomes even more demanding [22, 23]. Moreover, when each nanometer counts, the required wet chemical development also becomes extremely tricky. The slightest variation in temperature and timing affects the resulting 3D nanostructures so they vary even across the same substrate.

There are a few alternatives to EBL for grayscale nanolithography, for example, focused ion beam (FIB) milling [24] and focused electron beam-induced deposition (FEBID) [25]. Both methods allow direct patterning of 3D nanostructures without wet development. However, besides the cost and complexity of these methods, a few other drawbacks prevent them from being universally used: both methods are slow, the substrate inevitably gets contaminated, and single-nanometer 3D precision is still hard to achieve.

Thermal scanning probe lithography (t-SPL) is a novel alternative approach for grayscale nanolithography [26]. It is based on direct thermal resist sublimation by a heated scanning probe. This technology appeared at IBM Research Zürich in the framework of the Millipede memory project [27], which has spun into a new nanofabrication method. In 2014, former IBM researchers created a start-up called SwissLitho to commercialize this technology as the NanoFrazor (SwissLitho AG, Zurich, Switzerland).

The NanoFrazor’s hot tip ‘writes’ the structure line by line and retraces it in the cold regime like a high-speed AFM [28], immediately imaging the topography of the just-written pattern, as illustrated in Figure 2. The writing depth is controlled using the patented closed-loop lithography (CLL) procedure [29]. It modulates the force applied to the cantilever using the feedback from the immediate line retracing, detecting sub-nanometer deviations from the target patterning depth. This approach immediately compensates for minimal thermal drifts or any other change in the environment that can affect the patterning, without measuring any external parameters but the shape of the actual nanostructure, itself. CLL enables unprecedented vertical resolution of a single nanometer.

The reliability of grayscale t-SPL was demonstrated by ultra-precise patterning more than 1000 write fields of 10×10 μm^2 with a single tip. The resulting error of the

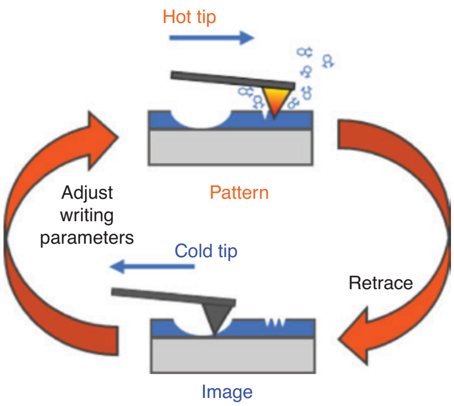

Figure 2: Schematic of the t-SPL working principle and Closed-Loop Lithography implemented in the Nanofrazor system: a hot tip creates the pattern by evaporating the PPA resist; then the cold tip retraces the same line imaging the resulting profile; patterning parameters are recalculated based on the collected depth information.

target depth was below 1 nm (1σ) for each of the fields. For comparison, such overall error is close to the best natural roughness of a spin-coated polymer film. The ultra-precise depth control can be maintained in shallow patterning regime (ca. up to 50-nm-deep features). Deeper than that, due to imaging limitations and other influences, the vertical resolution gets slightly worse. Overall patterning depth with current cantilever geometry and tip length can reach only around 150 nm. Large pattern amplification by reactive ion etching was shown in Silicon reaching more than 100×. The depth of up to 4 µm [30] was demonstrated while maintaining low roughness of the etched surface [31].

Compared to other lithography methods, t-SPL has a few very important advantages: it skips the wet development step, it does not damage or charge the substrate [32], and it does not require vacuum. The endothermic decomposition and evaporation of polyphtalaldehyde resist [PPA, most common resist used for the t-SPL, AR-P 8100 (Phoenix 81) from Allresist GmbH, Strausberg, Germany] is highly localized, so there are no proximity effects that need corrections. In this way, a lateral resolution below 25 nm is routinely achieved (record at 8 nm half-pitch). Besides, the total cost of NanoFrazor ownership is considerably smaller than that of a dedicated e-beam system, mainly because no UHV, no special shielding and room, and no high-voltage electron optics are required.

Early highlights of the unique ultra-precise grayscale capabilities of t-SPL are the Guinness world records for the smallest Matterhorn replica and a 3D world map, made shortly after the invention of the technology in 2012 at the IBM Research lab in Zürich [33]. Since its commercialization in 2014, the technology has already enabled several unique research applications that would have been impossible to make by any other means. A few recent examples are shown in Figure 3: photonic molecules [19], nanofluidic rocking Brownian motors [18],

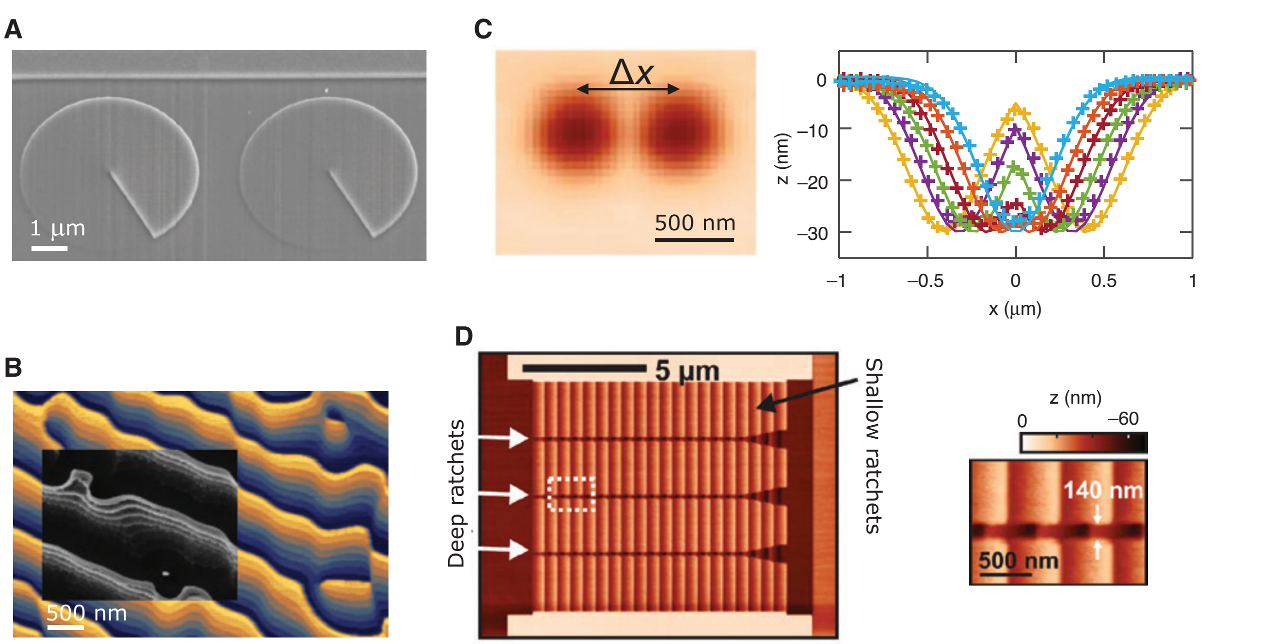

Figure 3: Examples of 3-dimensional nanostructures obtained by t-SPL using the NanoFrazor: (A) Spiral phase plate etched into Silicon; (B) 64 nm deep computer-generated hologram pattern; (C) Twin Gaussian cavities for a stack forming distributed Bragg reflectors, comprising a photonic molecule and average cross-scans taken with 160 nm step [3]; (D) Ratchet structure for the rocking Brownian motion nanofluidic device, the inset showing the outlined section [4].

and 3D phase plates for transmission electron microscopes [19].

t-SPL is not confined to academic research, though. The 3D nanostructures produced by the NanoFrazor can be precisely replicated by pattern transfer from PPA resist into soft nanoimprint stamps or molding into the UV nanoimprint resist. Such grayscale master molds were successfully replicated using Ormostamp from Microresist Technologies (micro resist technology GmbH, Berlin, Germany), the SCIL nanoimprint process, and the EVG SmartNIL™ process (EV Group, St. Florian am Inn, Austria) [34]. Also, PPA resist with a 3D nanoscale pattern was successfully electroplated with Ni. The resulting Ni shim was then used to replicate 3D nanostructures into PMMA by means of vario-thermal injection molding [35].

Currently, the NanoFrazor systems can only pattern using a single tip. This limits typical total patterning area at a high vertical resolution to about 100 000 μm^2 before the tip needs to be exchanged (which is a simple procedure, though). Larger throughput can be achieved in several ways. Multi-tip systems are already in development, targeting coverage of cm^2 areas without compromising the resolution. There already exists a working prototype for the 8-inch wafer fabrication, and a concept for a multi-head/multi-tip tool capable of patterning even larger areas is being developed. Another feasible approach is mix and match with other technologies, for example, with direct laser writing. The first laser writer was recently integrated with the NanoFrazor Explore system in collaboration with SwissLitho and Heidelberg Instruments. The hybrid system aims to increase the speed of the binary (2D) lithography, both the hot tip and the laser evaporating the resist at the corresponding nano- and microscale. A similar approach is envisioned to be extended to the grayscale mix and match direct-write lithography.

5 Conclusions

Grayscale lithography is widely used for different industrial and scientific applications. With further development of such applications, 3D micro- and nanostructure manufacturing will grow even more important in the future.

There are different approaches to microscale grayscale lithography, suitable for highly complex structures, and large-volume manufacturing, too. The direct laser write technology is well established (lateral resolution 300 nm, vertical resolution 50 nm), and new applications motivate further process optimization to satisfy stringent quality and throughput requirements. Development of binary (2D) direct laser write lithography will help to further improve the capabilities of grayscale technology. However, grayscale-specific challenges also arise (such as height limit of 60 μm), driving the development of new optical and computational solutions and suitable resists.

Thermal probe scanning lithography implemented by the NanoFrazor enables sub-10 nm lateral resolution and single-nanometer vertical resolution, thanks to the closed-loop lithography. This ultra-precise 3D capability has already enabled scientific breakthroughs and can also be used for the manufacturing of master molds for nanoimprint lithography or injection molding. In the future, it will be scaled up for industry-relevant throughput using multiple tip writing and mix and match with direct laser writing.

References

[1] M. Kneissl, in: 'III-Nitride Ultraviolet Emitters: Technology and Applications', Eds. By M. Kneissl and J. Rass (Springer International Publishing, Cham, Switzerland, 2016) pp. 1–25.
[2] W. Mönch, Adv. Opt. Technol. 4, 79–85 (2015).
[3] E. G. Loewen and E. Popov, Diffraction Gratings and Applications (CRC Press, Boca Raton, USA, 2018).
[4] F. Duerr, Y. Meuret and H. Thienpont, Appl. Opt. 49, 2339–2346 (2010).
[5] K. Shanks, S. Senthilarasu and T. K. Mallick, Renew. Sustain. Energy Rev. 60, 394–407 (2016).
[6] L. Wang, S. Kruk, H. Tang, T. Li, I. Kravchenko, et al. Optica 3, 1504–1505 (2016).
[7] N. Yu and F. Capasso, Nat. Mater. 13, 139–150 (2014).
[8] M. C. Traub, W. Longsine and V. N. Truskett, Annu. Rev. Chem. Biomol. Eng. 7, 583–604 (2016).
[9] C. Zhang, D. A. Mcadams and J. C. Grunlan, Adv. Mater. 28, 6292–6321 (2016).
[10] S. Klammt, A. Neyer and H. F. O. Müller, Sol. Energy 86, 1660–1666 (2012).
[11] Y. Lu and J. Cecil, Int. J. Adv. Manuf. Technol. 84, 1141–1152 (2016).
[12] A. Kumar, Manuf. Lett. 15, 122–125 (2018).
[13] D. Huo, K. Cheng and F. Wardle, Int. J. Adv. Manuf. Technol. 47, 867–877 (2010).
[14] P. Yao, G. J. Schneider, B. Miao, J. Murakowski and D. W. Prather, Appl. Phys. Lett. 85, 3920–3922 (2004).
[15] T. E. Dillon, A. Sure, J. A. Murakowski and D. W. Prather, J. MicroNanolithography MEMS MOEMS 3, 550–555 (2004).
[16] R. Houbertz, P. Declerck, S. Passinger, A. Ovsianikov, J. Serbin, et al. Phys. Status Solidi A 204, 3662–3675 (2007).
[17] L. H. Gabrielli, D. Liu, S. G. Johnson and M. Lipson, Nat. Commun. 3, 1217 (2012).
[18] M. J. Skaug, C. Schwemmer, S. Fringes, C. D. Rawlings and A. W. Knoll, Science 359, 1505–1508 (2018).
[19] C. D. Rawlings, M. Zientek, M. Spieser, D. Urbonas, T. Stöferle, et al. Sci. Rep. 7, 16502 (2017).
[20] B. Feng, J. Deng, B. Lu, C. Xu, Y. Wang, et al. Microelectron. Eng. 195, 139–144 (2018).
[21] S.-Y. Lee and K. Anbumony, J. Vac. Sci. Technol. B Microelectron. Nanometer Struct. Process. Meas. Phenom. 25, 2008–2012 (2007).

[22] Q. Dai, S.-Y. Lee, S.-H. Lee, B.-G. Kim and H.-K. Cho, J. Vac. Sci. Technol. B Nanotechnol. Microelectron. Mater. Process. Meas. Phenom. 30, 06F307 (2012).
[23] S. Pfirrmann, R. Kirchner, O. Lohse, V. A. Guzenko, A. Voigt, et al. “mr-PosEBR: a novel positive tone resist for high resolution electron beam lithography and 3D surface patterning,” presented at the SPIE Advanced Lithography, San Jose, California, United States, 2016, p. 977925.
[24] C.-S. Kim, S.-H. Ahn and D.-Y. Jang, Vacuum 86, 1014–1035 (2012).
[25] I. Utke, A. Luisier, P. Hoffmann, D. Laub and P. A. Buffat, Appl. Phys. Lett. 81, 3245–3247 (2002).
[26] A. W. Knoll, D. Pires, O. Coulembier, P. Dubois, J. L. Hedrick, et al. Adv. Mater. 22, 3361–3365 (2010).
[27] P. Vettiger, G. Cross, M. Despont, U. Drechsler, U. Durig, et al. IEEE Trans Nanotechnol. 1, 39–55 (2002).
[28] R. Garcia, A. W. Knoll and E. Riedo, Nat. Nanotechnol. 9, 577–587 (2014).
[29] A. W. Knoll, M. Zientek, L. L. Cheong, C. Rawlings, P. Paul, et al. Proceedings Volume 9049, Alternative Lithographic Technologies VI; 2014, 90490B.
[30] Y. Lisunova, M. Spieser, R. D. D. Juttin, F. Holzner and J. Brugger, Microelectron. Eng. 180, 20–24 (2017).
[31] Y. Lisunova and J. Brugger, Microelectron. Eng. 193, 23–27 (2018).
[32] X. Zheng, A. Calò, E. Albisetti, X. Liu, A. S. M. Alharbi, et al. Nat. Electron. 2, 17 (2019).
[33] “IBM scientists create the smallest 3D map of planet Earth.” [Online]. Available: https://phys.org/news/2012-01-ibm-scientists-smallest-3d-planet.html. [Accessed: 01-Apr-2019].
[34] T. S. Kulmala, C. D. Rawlings, M. Spieser, T. Glinsner, A. Schleunitz, et al. SPIE Proc. 10584, 1058412 (2018).
[35] C. Rytka, P. M. Kristiansen and A. Neyer, J. Micromech. Microeng. 25, 65008–65023 (2015).