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# **TOPICAL REVIEW**

# Massively parallel fabrication of repetitive nanostructures: nanolithography for nanoarrays

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#### Abstract

This topical review provides an overview of nanolithographic techniques for nanoarrays. Using patterning techniques such as lithography, normally we aim for a higher order architecture similarly to functional systems in *nature*. Inspired by the wealth of complexity in nature, these architectures are translated into technical devices, for example, found in integrated circuitry or other systems in which structural elements work as discrete building blocks in microdevices. Ordered artificial nanostructures (arrays of pillars, holes and wires) have shown particular properties and bring about the opportunity to modify and tune the device operation. Moreover, these nanostructures deliver new applications, for example, the nanoscale control of spin direction within a nanomagnet. Subsequently, we can look for applications where this unique property of the smallest manufactured element is repetitively used such as, for example with respect to spin, in nanopatterned magnetic media for data storage. These nanostructures are generally called *nanoarrays*. Most of these applications require massively parallel produced nanopatterns which can be directly realized by laser interference (areas up to  $4 \text{ cm}^2$  are easily achieved with a Lloyd's mirror set-up). In this topical review we will further highlight the application of laser interference as a tool for nanofabrication, its limitations and ultimate advantages towards a variety of devices including nanostructuring for photonic crystal devices, high resolution patterned media and surface modifications of medical implants. The unique properties of nanostructured surfaces have also found applications in biomedical nanoarrays used either for diagnostic or functional assays including catalytic reactions on chip. Bio-inspired templated nanoarrays will be presented in perspective to other massively parallel nanolithography techniques currently discussed in the scientific literature.

(Some figures in this article are in colour only in the electronic version)

# 1. Introduction

The vast number of publications concerning lithographic techniques, which are capable of forming structures with lateral dimensions below 100 nm, suggest that high-end exposure techniques are well established. Particularly, in microelectronic systems production machines are currently under development reaching the 32 nm node, which is

defined as the average half-pitch within a semiconductor pattern. Ultimate resolution, i.e. controlling functional elements at the molecule, atom, electron or spin scale remains the major driving force for further investigations in lithography. This is a research field that we currently name *nanolithography*. A selection of nanolithographic techniques, which are investigated for the massively parallel fabrication of repetitive nanostructures, is discussed in this review based



Figure 1. Definition of lithographically manufactured nanoarrays consisting of nanofeatures at the scale of 100 nm.

on the following two criteria: (1) capability of the lithographic technique to resolve lateral features at a scale below 500 nm and (2) providing at the same time an extremely high ordering of these features across the desired manufacturing area. Features containing these two properties are generally called *nanoarrays*.

A single feature reduced in its size down to the nanoscale (e.g. a dot) may by itself already possess remarkable properties, such as very unique properties of electron transport, yet to be discovered fully [1]. Applications, where the repetitive nature of uniform nanostructures across a large manufacturing area come into play, have already demonstrated their potentials, e.g. in patterned media (PM) for data storage [2]. This review is a compilation of the recent literature found with respect to the nanolithographic principles used in the pattern transfer of these massively parallel nanostructures. It is important to distinguish these nanoarrays with geometrical features at the nanoscale from biological screening arrays containing nanoliter sized wells, which are also called nanoarrays [3].

Often the research on nanolithography for the fabrication of a nanoarray is dedicated to a specific field of application, e.g. the fabrication of data storage media, photonic crystals and nanoarrays for diagnostics. For a better understanding of categorizing these lithographic techniques figure 1 shows the relation between the length scales of interest with its extreme demands on placement accuracy to gain the required high ordering of the nanofeatures. Ideally, nanoarray patterns can be produced across the full area of a standardized substrate, such as a silicon wafer with a diameter of 4 inch or even larger. However, for many devices such extreme large-area continuous patterning may not be required. Considering that defect-free patterning across the area of a typical microelectronic silicon die of  $26 \times 33 \text{ mm}^2$  is already a major challenge, we define here an area of 1 cm<sup>2</sup> as large-area patterning in the context of nanoarrays. Figure 2 presents two kinds of nanoarray layouts: the continuous and the discontinuous nanoarray, of which the latter is defined by repetitive clusters of nanoscale features that by themselves are also ordered. Comparing nanoarrays of a certain material with a continuous thin-film layer of the same material can deliver novel properties to the layer, of which one application example is a photonic crystal that represents tunable optical properties in guiding light within these nanostructured materials defined by the lateral features size of the individual nanostructures, their shape and periodicity. These nanostructured materials are often called meta-materials.

Finally, we need to define the difference between lithography and pattern transfer. Generally, transfer intrinsically means that a pattern is carried over from one layer to another. These layers are chosen for their masking, functional or sacrificial nature, such as for example a silicon nitride layer on silicon. The layer protects the silicon during etching in, e.g. potassium hydroxide (KOH).

Typically, lithography employs a resist layer. This layer is normally a polymeric layer being sensitive to light of a certain wavelength and being etch-resistant in the transfer step, hence the name resist. Depending on the choice of lithographic technique a pattern is transferred from a mask to a resist by an exposure step, from a resist to an underlying functional layer by etching or from a resist to an added layer by, e.g. deposition and lift-off. Many varieties of process sequences of pattern transfer are known and several extensive writings are available, of which one good introduction to this topic is, for example, *Handbook of Microlithography, Micromachining, and Microfabrication* edited by Rai-Choudhury [4].

However, at a length scale beyond the lateral width of 500 nm it becomes extremely difficult to fabricate structures from the top-down using ultraviolet (UV) photolithographic techniques through a mask, blocking the light in certain regions, because of defects due to diffraction of the light at the mask openings. It is already known and established in industry that moving to a shorter wavelength, specifically deep ultraviolet (DUV) is extremely well suited for the sub-500 nm domain. Unfortunately, only few such industrial systems are accessible to researchers in the field of nanotechnology. Since these machines are highly dedicated to electronics, these will not be reviewed in detail here but only briefly discussed in the context of next generation nanolithographic principles in section 2. Alternatively, any feature can be made quite reliably even down to a few nanometres by electron-beam lithography. Of course, current state-of-the-art electron-beam (e-beam) lithography is an indispensable technique concerning the fabrication of templates (also named masters or moulds). On the other hand, this classical top-down technique is extremely



**Figure 2.** Definition of (*a*) discontinuous and (*b*) continuous nanoarrays and (*c*) possible errors. Both (*a*) and (*b*) can deliver meta-material properties, which depend on the combination of the dimension of the single feature and the meta-period ( $P_{meta}$ ) of its repetition within the array. A field is defined here by  $1000 \times 1000 \text{ nm}^2$ , alignment errors, positioning from field to field (stitching), overlay errors and placement errors within one field can occur. These errors will depend on the lithographic technique and are not generally defined. A typical microelectronic chip (die) covers an area of  $26 \times 33 \text{ mm}^2$ . In semiconductor industry a roadmap is assembled to illustrate lithographic needs for innovation. A novel lithography technique must guarantee a certain degree of fault-free structuring within the patterning fields across the die. Of course, the exact definition of feature tolerances highly depends on the function of a device. For semiconductor devices a summary of these definitions can be found at [5].

slow and, referring to figure 2, this type of pattern delineation suffers in nanoarrays from stitching errors and discontinuities alike. Therefore, e-beam lithography also will not be further discussed in detail because of its serial write character, besides a very promising new particle beam lithography generation, utilizing so called multiple electron- or ion-beam sources.

The diversity of novel concepts utilizing templates for nanoarray fabrication will be discussed thoroughly. Specifically, in the field of medical applications a multitude of nanoarrays can be found for diagnostic purposes, however functional arrays exist, too, for example exploiting enzyme reactions on chip. An overview of these selfassembly techniques 'from the bottom-up', even exploring biological substrates such as DNA, is given in section 3. Subsequently, in section 4 we will discuss nanoimprinting. Nanoimprint lithography (NIL) is a top-down templateassisted nanolithographic method, which is already suitable for production standards yielding cost-effective pattern transfer exploiting the existing semiconductor mask standards for template fabrication. Figure 3 illustrates these two opposite approaches of nanotechnology, top-down and bottom-up, given by examples of their applications.

In the case of utilizing interference, the initial lithographic step can be performed without a mask. For specific applications, including relatively low production volumes, this can be an advantage. Although only briefly mentioned in section 5 the developments of maskless techniques such as interference lithography (IL), are considered important alternatives to the aforementioned photo-, e-beam, self-assembly and nanoimprint lithographic techniques, specifically for initial patterning of large-area periodic nanostructures.

Throughout the last two decades, there has been a diversity of new resist systems in lithography and the stringent demands for innovation in the semiconductor industry have fostered tremendous technical improvements concerning exposure tools. When setting up a new exposure tool it initially requires the definition of a process line, specifically with respect to nanolithography. As one such example, this report includes IL nanofabrication as it has been utilized at the MESA+ Nanolab. Periodic nanostructures are achieved in a single, maskless lithography step, and for example, recording experiments in a diversity of specifically designed PM materials including the development of read/write technology at the nanoscale can readily take place. Beyond this specific application our laser IL (LIL) system has also been explored for a variety of experiments aiming at the study of photonic crystals, hierarchical nanostructures by self-organized (templated) assembly and the creation of nanostructured surfaces for the investigation of a novel class of nanotopologically designed biomaterials. These approaches will be discussed in section 6.

In sum, this topical review presents the technological aspects of a selection of nanolithographic processes for nanoarrays. It may also offer a starting point for further research on design rules for massively parallel nanofabrication. This review discusses the broad field



**Figure 3.** Top–down and bottom nanotechnology. In nanolithographic methods, generating structures at the nanoscale from the top–down maskless or through a mask, patterning can be achieved by writing or printing (from a mask or mould). When a mould is used for imprinting the technique is sometimes also called stamping. In nanotechnology from the bottom-up by self-organization of species, such as molecules and solid-state architectures (nanocrystals, quantum dots) from the atomic to the mesoscopic scale, we can distinguish either pure self-assembly or a combination thereof with a predefined template generated from the top–down. If such a combination is used we often also call these techniques nanolithography. Shown (clockwise from top) are an electron microscopy image of a nanomechanical electrometer obtained by e-beam (top–down), patterned film of carbon nanotubes obtained by microcontact printing and catalytic growth (template-assisted bottom-up), a single carbon nanotube connecting two electrodes (electrodes by top–down, whereas positional assembly of the carbon nanotube onto the electrodes is performed manually utilizing the tip of a scanning probe), a regular metal-organic nanoporous network integrating iron atoms and functional molecules, whereas the iron atoms act as a template in the coordination of the molecules (self-organization from the bottom-up, sometimes also called coordination chemistry), and seven carbon monoxide molecules forming the letter 'C' positioned with the tip of a scanning tunnelling microscope. (Figure reprinted from [6] by permission from Macmillan Publishers Ltd.: Nature © 2002.)

of available nanolithographic routes for the fabrication of nanoarrays. Depending on the size and shape of the individual nanostructure and the requirements of the extent of periodicity within the nanoarray, a nanolithographic fabrication technology must be chosen. The selection of a specific nanolithographic technique will also strongly depend on the field of application. Various examples of nanoarray applications in information storage and medicine are presented.

# 2. Nanolithographic techniques

The sequence of process steps involved in fabricating a masking layer and allowing the carry over of a pattern from the mask into a functional layer of material is called lithography. Figure 4 depicts the four lithographic principle utilizing a masking layer being patterned through either lithographic exposure (see figure 4(a) and (b)) or imprinting (see figure 4(c)) via a photomask or template, respectively, in comparison with the patterning of a masking layer by self-assembly (see figure 4(d)). The actual transfer into the functional material by deposition, lift-off, etching or else, e.g. layer-by-layer

deposition as described by Hua *et al* is, generally speaking, not part of the lithographic sequence [7].

This definition has been weakened in the explorations of novel nanostructuring methods, such as scanning probe lithography that directly produce a functional nanostructure, e.g. dip-pen lithography. Several outstanding contributions have been made in the field of direct-write scanning probe lithography techniques by the Mirkin Research Group at the Northwestern University. These researchers utilize multiple cantilevers fabricated by microelectromechanical system's technology to increase throughput. One of their specific focal areas is controlling architectures on the 1-100 nm length scale, demonstrating dip-pen massively parallel pattern transfer in nanomedicine, examples of which are described elsewhere [8–11]. However, categorizing any of these lithographic techniques in terms of their importance for the field of miniaturization is not possible. According to figure 4, it is much more vital to be able to conclude for a certain application on the question if a masking or templating layer is needed, and if so, the choice of a specific transfer process gains certain benefits in respect of the realization of the functional properties of nanoarrays. Of course, this conclusion may



**Figure 4.** Principles of lithographic methods. Lithography utilizes in its strict definition a masking layer. A variety of technologies exist to generate such masking layer. In most cases the mask or template fabrication process by itself is also a lithographic process using a direct-write method, mostly e-beam lithography. Although stencil masks, e.g. fabricated by techniques such as focused ion-milling are possible, they are not broadly established in manufacturing. The most established technique is (*a*) photolithography through a so called chromium mask. Due to the resolution limitations in photolithography often nanofeatures at the nanometre scale are transferred by nanoimprinting either utilizing a wet-process, hardening a polymeric layer through (*b*) UV exposure or by the application of a (*c*) thermal process pressing a template into a thermoplastic layer. Alternatively, a masking layer can be generated by (*d*) self-assembly, whereas in some of these self-assembly nanolithographic techniques the spacer or template molecule can be omitted. The common aspect of lithographic techniques is defined here as the fact that the pattern generated in the masking layer is subsequently transferred into a functional material.

be valid for many other designs, too. Here, however, the review is further concerned with nanofabrication techniques for massively parallel pattern transfer yielding nanoarrays.

#### 2.1. Top-down nanolithographic principles

From all existing top-down nanolithographic methods in practice the e-beam direct-write approach offers the highest pattern fidelity (freedom of shape, accuracy and precision of features and overlay). Because of its slow nature it is mainly used to produce masks and template structures. In combination with nanoimprinting it provides a versatile and cost-effective route for the fabrication of many interesting So far, this serial write technique cannot nanodevices. be replaced for template manufacture. Although LIL is a much faster and cheaper method to be utilized as a maskless massively parallel patterning technique at the initial patterning step, unfortunately, it cannot compete with e-beam lithography in wafer-scale pattern uniformity and freedom of pattern geometry. These drawbacks of LIL may be limited in the development of suitable large-area mirrors or other suitable interference optics, however, 193 nm DUV and extreme ultraviolet (EUV) stepper-and scanner techniques already belong to the emerging lithographic technologies for the semiconductor industry offering a very high precision and resolution. Since these technologies are mainly restricted to very high volume production only a brief overview is given. Work on these new light sources was initiated amongst others by contributions of Bokor and Oldham [12, 13]. Maskless techniques including multiple electron/ion-beam lithography and maskless DUV/EUV IL may at some point overcome the limitation of low throughput in e-beam lithography at least for specific periodic structures. To overrule any of the established photolithographic methods, however, the selection of a nanolithographic technique in manufacturing will be always a search for a compromise between machine cost, speed (wafer throughput) and pattern fidelity (resolution and process stability). In high volume production, where throughput is the major argument for choosing a specific lithography technique, template replication processes may be a successor of the classical photolithographic step; even so the initial costs in making a template can be high. Although of much interest to industry, at this moment, the topic of defining an appropriate cost model of nanodevices manufactured based on either one of the nanolithographic strategies goes beyond the intention of this review.

A totally different template-free approach for nanoarray fabrication is worth mentioning. Materials can be manipulated by low-dose electron beams. For example, by exposing a thinfilm polymer to the transmission electron microscope real-time direct manipulation of the polymer chain aggregate structure in two dimensions becomes possible, melting the polymer with subsequent movement and attraction towards preferred electron concentrations within the beam. This discovery is still highly experimental and has been described by Brown et al and the principle may be compared with other direct energetic beam-induced surface tension gradients yielding the formation of patterns in material by, e.g. laser interference metallurgy [14, 15]. Before we summarize in the next section the various techniques for templated self-organized nanoarrays contemplated from the recent scientific literature, this review also gives a brief overview of the various emerging topdown lithographic principles being explored with the aim of next generation industrial nanodevice fabrication in the following subsections. Besides the development of new machine architectures also novel resists and etch-through processes for lithographic pattern transfer are developed, too, as for example recently reviewed by Argitis et al [16]. For more recent examples for finding the novelty of such processing details Proceedings of SPIE is one of the best sources, e.g. volume 6923 (2008) Advances in resist materials and processing technology XXV. Therefore novel lithographic resist systems will not be further referred to in this review.

#### 2.2. Emerging lithographic technologies in industry

2.2.1. DUV lithography is a key enabling technology using 193 nm excimer lasers to produce commercial chips at the 90 nm technology node. Validation of the 193 nm immersion scanner at IMEC's 300 mm wafer fab facility in Leuven, Belgium, is a clear milestone towards ensuring the availability of a cost-effective technology at 65 nm and may even be extended to the 45 nm node for microelectronics. Many

publications have been presented concerning this technological development, of which only a few to name are [17–21]. The development consists of the definition of a suitable light source, optics, and other hardware aspects of such a lithographic system, e.g. including mask technology, immersion liquid and defect analysis [22].

2.2.2. EUV lithography is just like DUV considered as one of the key enabling technologies. The technique is currently mainly investigated to fill the gap in the semiconductor roadmap towards the 22 nm node for production. At the moment when the technology roadmap gets to its absolute limit of the photolithographic method, however, an upcoming trend is the utilization of the 'More than Moore' concept [23–27]. Therefore industry's dedication to EUV as a next generation lithography tool is focused on by very few players [28–33]. At very dedicated research centres and lithography tool manufacturers, this development of EUV technology takes place. IMEC in Europe and the Lawrence Berkeley National Laboratory in CA, USA, are leading the EUV development. Reaching already towards the 32 nm node generation of very large-scale integrated (VLSI) electronics by optical lithography utilizing a 248 nm laser source, EUV is still one of the top candidates for industry to adopt in production. Although the investment for such machinery is very high the EUV technique offers very reliable access to high resolution nanostructures, which will be clearly able to satisfy the market needs for high volume nanoelectronic device production for some additional decades.

2.2.3. Multiple e-beam/ion-beam lithography is a rather different approach to the developments of DUV and EUV scanners. It follows the idea of upgrading the wafer throughput of direct e-beam/ion-beam lithography by controlling multiple beams in parallel across the wafer area [34–42]. These maskless, massively parallel patterning techniques aim for critical dimensions at the 50 nm line width. Several researchers have demonstrated the potential of such lithography concept by fabricating multiple-beamlet arrays utilizing MEMS technology. Yet, it is not possible to estimate the position of these emerging technologies on the industrial roadmap.

# 3. Templated self-organized nanoarrays

Within medical diagnostics label-free and high resolution protein/DNA nanoarrays are an important development. Sinensky and Belcher described a novel type of such arrays using Kelvin probe force microscopy as a detection technique [43]. With their method, probing surface bound target biomolecules, they show that nanoarray diagnostics offering a 10 nm resolution is potentially possible. Although there is no elaboration of nanolithographic fabrication strategies mentioned in their paper, and their choice of dip-pen nanolithography (DPN) over other nanolithography techniques may be arbitrary, it is a rather interesting question, what the specific requirements for this class of nanoarrays are. The utilization of one of the templating techniques as described as follows may appear highly suitable, Topical Review

too, in this field of applications. Templated nanoarrays may be achieved by both the top–down or bottom–up nanofabrication concepts. In this section the focus is on the self-ordering of molecules or particles that form either a templating layer or even a three-dimensional scaffold for the subsequent pattern transfer into the functional material of the nanoarray. Reaching the resolution limit of physical templating by top–down lithographic techniques many other substrates including biological ones are also explored. In the following subsections some specifically interesting techniques emerging from the expanding knowledge of self-assembly are described.

# 3.1. Bio-based nanofabrication

3.1.1. Virus-based templating is one approach for the fabrication of nanoarrays. Lee *et al* for example, stated in their publication in Science the ordering of quantum dots using genetically engineered viruses by fabricating highly ordered composite materials [44]. In their paper highly ordered A7-ZnS self supporting viral films are introduced of which figure 5 gives an overview of the involved engineering length scales of such films. This example demonstrates the extent of possibilities by self-assembly even anticipating the request as given previously in figure 1. A similar process is presented by Huang *et al* who demonstrated programmable assembly of nanoarchitecture also using genetically engineered viruses [45].

3.1.2. DNA-based templating may be used similarly as the aformentioned phages to introduce ordered patterns. In 2006, Feldkamp et al compiled a review on rational design for DNA nanoarchitectures, whereas DNA is used as a building block at the nanometre length scale. This is a technology concept, which can offer a large variety of designs for self-assembled supramolecular arrays and other scaffolds, which are possible to form bio-synthetic, mechanical and logical nanodevices. To avoid replication of data from Feldkamp's review, here, we will limit the overview of fabrication principles based on biological templating to those DNA-based techniques that have been applied to form highly ordered nanoarrays and are described in the literature over the last two years. For example, Becerril et al have reported on the use of DNA-templates for nickel nanostructures [46]. These nickel nanomaterials may be applied directly as catalytic platforms or for the selective binding of protein assemblies, which have been demonstrated for the reversible association of histidine-tagged phosducinlike protein. Another example of the exploitation of the inherent recognition and self-assembly capabilities of DNA is described by Wang et al [47]. The researchers synthesize and characterize a matrix assembly of single-wall carbon nanotubes (SWNTs) functionalized with carboxylic groups and single-stranded DNA (ssDNA) demonstrating negative differential resistance in the biotemplated interfaces of the matrix structure. Combinations of this bio-coordination chemistry will allow for a diversity of nanoelectronic effects and have yet to be investigated. Chhabra et al discuss a spatially addressable multiprotein nanoarray based on



**Figure 5.** Characterization of A7-ZnS film. (*a*) Photograph of A7-ZnS viral film. (*b*) POM (20) birefringent dark and bright band patterns (periodic length 72.8  $\mu$ m) were observed. These band patterns are optically active, and their patterns reverse depending on the angles between polarizer and analyzer. (*c*) Photoluminescent image, with an excitation wavelength of 350 nm and with filtering below 400 nm, shows 1  $\mu$ m stripe patterns (50). (*d*) SEM images of highly packed three-dimensional bulk film structure. (*e*) Schematic structural diagram of the A7-ZnS composite film. (*f*) AFM image of the free surface. The phage forms parallel aligned herringbone patterns that have almost right angles between the adjacent director (arrows). (*g*) SEM image showing the close-packed lamellar structure of phage and nanocrystal layers (red arrows) in the inner areas of the film. (*h*) Low-resolution TEM image of cross section of A7-ZnS film, with 20 nm × 2  $\mu$ m ZnS nanocrystal stripe pattern aligned between one phage length in the *x*-*z* direction of film; the inset shows an electron diffraction pattern of ZnS wurtzite structure. (*i*) Low-resolution TEM image of film viewed in the *y*-*z* direction, showing ZnS nanocrystals. (Figure reprinted from [44] by permission from the American Association for the Advancement of Science © 2002.)

aptamer-tagged DNA templated nanoarchitectures [48]. Han Yan's group and collaborators of the Arizona State University published around 50 papers dedicated to DNA-tile-based self-assembly and its application to biosensors. One of these contributions is particularly worth mentioning among the recent developments of nanotechnology describing the phenomenon of multivalency, which is suitable to assemble multiple molecules while controlling the spacing between Finally, it is of interest to mention in this them [49]. context that proteins in general can be utilized to produce A fairly recent attempt by Allred et al nanoarrays. describes the three-dimensional inorganic nanoarchitectures electrodeposited through a surface(S)-layer protein mask [50]. Unfortunately, the full scale of these fascinating applications of nanoarrays and their fabrication strategies cannot be discussed in this review; however, further literature giving an extensive overview of this technology can be found easily as, for example, a description of recognition imaging and highly ordered molecular templating of bacterial S-layer nanoarrays containing affinity tags by Tang et al [51].

#### 3.2. Guided self-assembly

Besides the use of biomolecules for templating and functioning in nanoarrays, geometrically constrained inorganic templates have been explored for devices, too. For example, nanofabrication of plasmonic structures was investigated by Henzie *et al* who describe the gain of some exceptional, optical properties due to multiscale patterning of metamaterials utilizing soft lithography [52]. Nonetheless an interesting class of devices, we will skip reviewing the fabrication of plasmonic devices in detail and complete this overview by recently published nanoarray fabrication techniques applying self-organization by itself or assisted by top–down fabrication methodology.

*3.2.1. Directed and chemical contrast-assisted methods* belong to the group of already established fabrication methods for bio-nanoarrays, which include the electrochemically controlled assembly of DNA and protein biomolecules onto gold nanoelectrodes and templating protein assemblies on micro-contact printed surfaces. Huang *et al* describe

a dynamically configurable biomolecular array adjusting the electrochemical potential applied at the nanoelectrode allowing for a variety of combinatorial interactions [53]. Further Iversen et al describe a functional nanoarray exploring SNAP-tag proteins affinity bound to patterns of streptavidin modified with BG-biotin that has been stamped onto a surface by micro contact printing (microCP) [54]. Also electrical-field assisted nanofabrication can be utilized to form multilayered structures composed of biomolecule-derivatized nanoparticles. In the example by Dehlinger *et al* a microelectrode-array device facilitates the rapid parallel electrophoretic transport and binding of biotin and streptavidin fluorescent nanoparticles to specific sites on the microarray [55]. A record of the production of nanopatterns by a combination of e-beam lithography and self-assembled monolayers for an antibody nanoarray is presented by Zhang et al [56]. Further, Chen and Li contemplated a record of nanotechnology: moving from microarrays towards nanoarrays [57]. In this context also, a review of the development of high-throughput methods for functional analysis of diatom genes is given by Montsant et al, whereas Lynch et al addressed functional protein nanoarrays for biomarker profiling [58, 59]. Parallel arrays of geometric nanowells controlling lateral dispersion of molecules have been recently described by Visnapuu et al [60]. In their paper, a process is demonstrated that allows the assembly and visualization of curtains comprising thousands of individual DNA molecules organized at a diffusion barrier on a lipid bilayer-coated surface by a combination of a geometric barrier pattern with nanofabrication utilizing single-molecule optical microscopy.

3.2.2. Block copolymers and colloidial materials can also be used to form nanoscale structures. Block copolymer deposition generally behaves in its self-organized ordering similar to submicrometre inorganic colloidal self-assembly on surfaces [61, 62]. Colloidal self-assembly had been used also in early applications of massively parallel pattern transfer, which is then called colloidal, particle, block copolymer or nanosphere lithography depending on the specific materials being used for providing a masking layer [63, 64]. However, the self-assembly of a thin film of, for example, a sphericalmorphology block copolymer is also mentioned as block copolymer micelle nanolithography and it can be specifically templated using an array of nanoscale topographical elements that act as surrogates for the minority domains of the block copolymer. This technique has been demonstrated by several authors and is also called guided self-assembly or graphoepitaxy [65-71]. Some remarkable results on the discrete patterning capability of directed assembly of block copolymer blends were demonstrated by Stoykovich et al showing nearly perfect alignment of the molecules around corner-shaped features [72]. A year later the possibility of more complex designs has been extended by Wilmes et al who explored bending of soft block copolymer nanostructures by lithographically directed assembly and give some more insight into the energy model behind the assembly process [73]. Although it is a straightforward and fairly simple to use nanopatterning technique with a large variety of design

parameters the control of long-range order and defects are experienced as a major challenge applying this technique in real-world devices [74]. We also investigate guided selfassembly at the MESA+ Institute for Nanotechnology utilizing block copolymer, nanocrystals and supramolecular systems deposited onto templates fabricated by lithography, such as LIL. This topic will be further discussed, including other applications of LIL, in section 6 describing some of the research in our own facility. Confined synthesis can generally be considered a novel scheme in chemistry. As a final example for novel templated techniques utilizing colloidial materials, therefore, we should not fail to mention confined synthesis recently discussed by Fan et al [75] Fan et al demonstrated hierarchical nanofabrication of microporous crystals with ordered mesoporosity that can be realized through confined growth within a three-dimensionally ordered mesoporous carbon, synthesized by replication of colloidal crystals composed of size-tunable silica beads of about 10 to 40 nm diameter. Moreover, the authors revealed novel crystal morphologies, consisting of faceted crystal outgrowths from primary crystalline particles, thus providing new insight into constricted crystal growth mechanisms underlying confined synthesis.

#### 3.3. Specifically pore-forming materials

3.3.1. Porous alumina has been utilized as one of the first physical templates for nanoarray applications. Anodized aluminium oxide (AAO) contains self-organized hexagonally ordered pores. This type of porous alumina, better called AAO-templates, is formed by anodizing aluminium in a suitable electrolyte usually sulfuric ( $H_2SO_4$ ), oxalic ( $H_2C_2O_4$ ) or phosphoric  $(H_3PO_4)$  acids [76]. Utilizing AAO-templates in nanofabrication has been one of the first techniques being explored to demonstrate particular nanoscale effects and collecting the according insight of nanoscale features specifically in the area of patterned magnetic media [77]. Here, this technology is described in respect of an even more complex structure. Hierarchical structured nickel nanoring and hollow sphere arrays were generated by morphology inheritance based on an ordered through-pore template and electrodeposition by Duan *et al* [78]. Dipping an alumina template (AAO) into colloid solution forms a colloidal monolayer with The formation of such fine structures on the nanoscale. morphologies is attributed to the preferential growth along the inner wall of the alumina pores, while the nanoflakelet fine structure originates from a morphology inheritance process of the transitional product Ni(OH)<sub>2</sub>, which leads to the final nanostructured Ni crystals showing good magnetic properties. These structures may find application in the fields of catalysis, magnetics, optoelectrics, surface-enhanced Raman scattering (SERS) and other new nanodevices. Already for some time the self-organized structure of alumina has been utilized for template formation. Her et al contribute to this field of research by demonstrating the fabrication of nanostructured titania. Their paper describes the formation of thin-film titania by spin coating in the sol-gel state and subsequent nanoimprinting using an alumina template to yield a dense

array of nanopoles. Nanopores in titania were as well achieved by utilizing a suitable mould structure [79]. These nanoarrays may also be useful for solar cell, photocatalytic and sensing applications. When it comes to bioapplications of AAO-templated nanostructures it has been demonstrated by Kim *et al* that fibroblast cells respond to the nanotopology of, e.g. polymeric surfaces (PMMA) replicated by AAO nanoimprinting [80]. This area of research is also further studied amongst other research groups by us investigating osteoblast response on large-area scalable nanotopology for a variety of biomaterials.

3.3.2. Carbon-based templates are similar to AAO exploited to guide the assembly process of the functional material of a nanoarray. A large variety of material combinations has been investigated for their ability to result in ordered, selforganized nanodot arrays. One of these example is given by Meletis and Jiang [81]. Their paper describes the formation of cobalt nanodots in diamond-like carbon thin films that have been deposited by magnetron sputtering in a plasma-assisted Ar/CH<sub>4</sub> discharge. Cobalt dots of 5 nm diameter, separated by 1–2 nm DLC boundaries and arranged in hexagonal arrays were produced on silicon substrates demonstrating ultrahigh resolution. Such well-ordered magnetic nanodots based on self-organization are promising for next generation patterned magnetic media.

3.3.3. Porous gels can be used similarly as porous inorganic materials as templates. One route being explored extensively in self-assembly is the utilization of pore-like structures produced by organic synthesis, of which an overview is given by Hirst et al [82]. They describe that specific gelator molecules can be assembled for functional nanoscale architectures. Synthetic chemistry exploiting this specific type of supramolecular structures that form molecular gels can be applied as nanostructured scaffold, e.g. for the regrowth of nerve cells, but has also gained interest in nanoelectronics. Redefining the limits of biomedical devices utilizing this type of smart polymeric gels has also been described by Chaterji et al who summarized the recent progress in the development and application of polymeric gels based on defining their properties, describing stimuli response and illustrating applications of smartly designed gels to microfluidics [83].

# 4. Nanoimprint technology

Returning to the establishment of a massively parallel pattern transfer technique suitable for industry some very good papers have been published concerning the developments and applications of NIL (see principle in figures 4(b) and (c)). Evolved from hot-embossing at the microscale, thermoplastic NIL is the earliest NIL technique developed by Chou's group [84–86]. In the context of this review, recently Morton *et al* demonstrated wafer-scale patterning of sub-40 nm diameter and high aspect ratio (>50:1) silicon pillar arrays by nanoimprint and etching [87]. Further developments in this area of research led to a combination of imprinting

and UV exposure, so called step-and-flash imprinting. This process has been keenly investigated and brought to industrial standards mainly by contributions from Willson (University of Texas, USA) and Resnick (Molecular Imprints, Inc.) who jointly published more than 20 papers concerning step-and-flash imprint lithography. One recent example referring to UV nanoimprint lithography for manufacturing applications is given by Choi *et al* [88]. Willson's group mainly continues to develop and optimize materials and processes for this technology [89]. Although these techniques are at the frontier of nanopatterning including hybrid techniques of, e.g. physical-confined colloidal lithography, here we will only briefly summarize some special applications of nanoimprinting concerned with arrayed features in applications.

#### 4.1. Patterned magnetic media

Before we start describing the specific nanoimprint processes let us recap some background on patterned magnetic media and the impact of nanoscale in this field of research. Original research on pattern media started in the late 1980s and early 1990s, of which record is given, for example, by Chou et al and New et al [90, 91]. Recently, researchers at the Hitachi San Jose Research Center, CA, beautifully presented a cover article concerning patterning on hard disk drives, which further confirms the industrial attractiveness of such systems [92]. Given the fact that this vision was presented just about two years ago whereas the first mention of perpendicular patterned media substrates for data storage was published by us and other researchers more than ten years ago, we can conclude that technological development needs a sufficient transit time from the research laboratory to industry including the development of robust fabrication technologies [93–95].

Figure 6 illustrates the concept of patterned media (PM) with single domain magnetic islands against the classical multiple grain media, which suggests solving the thermal stability problem using a predefined magnetic dot per bit [96]. Realizing dots at a size of 25 nm diameter and periodicity of 35 nm, respectively, within a cost-effective high volume production process of PM disks is still a major challenge in nanofabrication. Using UV-curing compounds in NIL is one of the emerging technologies showing the capabilities in achieving both the high required throughput and resolution for this class of devices.

Previously a review by Moritz *et al* presented PM disk fabrication [97]. They demonstrated a fabrication method towards ultrahigh-density magnetic recording by the utilization of pre-etched wafers comparing the two ways of patterning (a) direct e-beam lithography and subsequent reactive ion etching and (b) nanoimprinting, where the nanoimprint technology delivers polymer arrays with features as small as 30 nm with a pitch of 60 nm, which are subsequently coated with a Co–Pt multilayer with perpendicular magnetic anisotropy. In 2005, Terris and Thomson reviewed in this journal self-assembled PM technology, which also pursues AAO-templates for the self-assembly of patterned magentic media [77]. The fabrication of high resolution PM also requires the development of novel read/write technology. One such



**Figure 6.** Overview of granular versus the PM for data storage. Comparing granular media with PMs, the novel concept consists of an array of magnetic islands created in the form of a dot. One dot represents a single domain, i.e. spin direction. One dot is either 'written' or 'non-written'. In the creation of spatially separated magnetic islands a higher resolution and lower noise level can be achieved than in conventional media. (Figure reprinted from [96] with permission from IEEE © 2008.)

approach can be found again in MEMS technology based on a cantilever design for atomic force microscopy originally introduced by Rohrer and Binnig [98,99]. More recent examples of patterned media are given in a review by Terris *et al* who also refer to nanoimprinting as one of the emerging routes for cost-effective nanofabrication of PM disks [100]. However, the authors do not give one particular process the benefit of surpassing the cost of introduction of this class of devices.

#### 4.2. Random access memory

A 6 nm half-pitch lines and  $0.04 \text{ mm}^2$  static random access memory (SRAM) pattern by NIL has been demonstrated by Austin *et al* [101]. The authors manufactured a specific high resolution mould by cleavage of a GaAs superlattice grown by molecular beam epitaxy. With these types of patterns they reached the limit of reliable SEM inspection; however, it is suggested that the UV curable nanoimprint process will even support smaller line width than 6 nm. Besides lateral resolution, one has to bear in mind that for faithful pattern transfer in nanoimprinting the remaining residual layer thickness is an important parameter due to the mechanicaldisplacement nature of this technique [102–104].

#### 4.3. Surface-acoustic-wave (SAW) devices

Researchers at the Paul-Drude Institut für Festkörperelektronik, Berlin, Germany, fabricated SAW transducers operating in the GHz range on LiNbO<sub>3</sub> using thermal imprint lithography [105]. The width of the metal lines was 170 nm at a period of 600 nm. Although these SAW devices do not challenge the resolution limit of this technology, which is currently in the order of 10 nm, they are an interesting example of devices having large volume applications in telecommunication.

# 5. Interference lithography

IL, of which the concept is illustrated in figure 7, can be carried out with any type of coherent light source; however, it becomes most interesting if a wavelength is used that reaches pattern periods below the resolution limit of the established mask pattern transfer utilizing an optical photolithographic aligner at  $\lambda = 365$  nm, e.g. by applying a DUV source and a Lloyd's mirror.

# 5.1. DUV interference nanolithography

Schattenburg et al demonstrated already in 1995 nanostructure fabrication by utilizing an argon-ion laser source at  $\lambda = 351.1 \text{ nm}$  [106]. The authors suggested interferometric lithography as a tool of choice for patterning large-area, phase-coherent periodic structures with submicrometre-period gratings and grids having found many applications including x-ray and EUV transmission gratings, EUV-UV polarizers and filters, field emitter arrays for flat panel displays, as well as in reference grids for e-beam lithography. An example of using this type of nanolithography of submicrometre sparse hole arrays for field-emission display applications is given by Chen et al [107]. This review presents multiple exposure at two different pitches (i.e. angle of incidence), the integration of interferometric and optical lithography by Moire alignment techniques; and various multi beam techniques that both provide a sparse array and result in a two-dimensional pattern in a single exposure. Later, the same



**Figure 7.** Artist impression of LIL for the fabrication of PM. The pattern period (P) is defined by the wavelength (lambda) and the angle of incidence (theta) while the possibility to rotate the substrate allows two-dimensional arrays such as dots, hexagones and Moiré-type patterns through a multi-exposure at different substrate rotations (alpha). LIL has been applied to fabricate the magnetic structured shown here in the inset, which is described in detail in [126].

group presents in an overview, next to optical lithography, interferometric lithography clearly as a nanotechnology enabler, demonstrating a large variety of applications, but specifically for large-area samples [108]. Several more publications concerning advanced maskless lithography even for the writing of arbitrary non-periodic patterns have been presented by researchers from the Bell Labs (today belonging to Alcatel Lucent, USA) [109–111]. Another more recent example of the utilization of interference in combination with catalytic etching is given by Choi *et al* [112].

#### 5.2. EUV interference nanolithography

Solak presented a striking review on this technique producing periodic nanostructures with resolution below the 20 nm [113]. Earlier work by Choksi *et al* introduced the concept of maskless EUV lithography [114]. Olav Solgaard and co-workers at Stanford University continued these developments publishing on analytical models for several micromirror array configurations and later on spatial light modulators for maskless lithography [115, 116]. More recent progress in EUV interferometric and holographic lithography are given by Cerrina's group at the University of Wisconsin-Madison, e.g. [117–119].

#### 5.3. Atom lithography

Laser focusing of chromium atoms for nanostructure fabrication was first described by McClelland et al [120]. This work was followed up by a publication on a fast, deterministic source of single chromium atoms [121]. So far, this topic is only very slowly progressing in the scientific community with one additional publication on the mechanism of an ultra thin coherent atom beam by Stern–Gerlach interferometry [122].

# 6. LIL at MESA+

A Lloyd's mirror laser set-up utilizing a cost-efficient 266 nm DUV laser is a simple and useful tool for the fabrication of nanoarrays receiving structural resolution down to the sub-50 nm regime, e.g. exploring combinations with reactive ion etching using the LIL resist as an etch mask. The application of cost-effective LIL has been previously demonstrated by us for the generation of sieves, photonic crystal structures and patterned magnetic media (PM) [123–127]. Figure 7 shows the concept for LIL exposure and an example of a resulting patterned media in the inset.

Based on earlier research by Cheng et al who investigated LIL templates for the fabrication of nanostructures with long-range order using block copolymer lithography, our set-up has also been applied for such hybrid lithographic techniques [128]. We investigated the patterning of surfaces by LIL assisting in the symmetry control of polymer colloidal monolayers and crystals formed by electrophoretic deposition [129]. Similar processes utilizing LIL were demonstrated also by other groups, e.g. Kim et al [130]. Additionally, our LIL system was investigated by us for preliminary templating experiments in layer-by-layer deposition of supramolecular assemblies. In the course of research concerning this hybrid approach, however, LIL has been replaced mainly by NIL techniques based on e-beam lithographically produced masters [131].

A variety of current investigations of nanostructure applications continue to rely on LIL as a nanolithographic tool at MESA+. Some record of these developments is mentioned in the following section.

# 6.1. Investigating 266 nm DUV Lloyd's mirror interferometric nanolithography utilizing multi-exposure and novel resist systems

A full record of this fabrication technology exploring positive chemical amplified resist (p-CAR) has already been given [132]. However, this review presents some additional results being achieved in the nanofabrication of large-area nanostructured surfaces exploring the capabilities of specific resists and multi-exposure. The method and materials are briefly summarized in the next section.

6.1.1. Method/materials for the generation of one-dimensional (gratings), two-dimensional (arrays of dots) and mesoscale quasi-regular discontinuous nanoarrays that were fabricated by the LIL exposure into resist are described briefly as follows. Standard (100) p-type, polished prime silicon wafers were used as substrates. LIL is operated at 266 nm utilizing the aforementioned continuous-wave laser source as an exposure tool. This lithographic process line consisting of spin coating, soft-bake, exposure, post-exposure-bake and wet development, has been described by us in detail earlier [132]. Three types of resist were employed. At first, a standard i-line photolithographic resist system OiR 907-13 (from Fujifilm, formerly called Olin) has been diluted such that it is suitable



**Figure 8.** Four multiple exposures resulting in a spatially defined pattern of micrometre and nanometre sized elements within a quasi-regular array, previously also defined as a discontinuous nanoarray. The mesoscale pattern can be uniformly fabricated across an area of approximately  $2 \times 2 \text{ cm}^2$ , the scale bar in the figure is  $1 \mu \text{m}$ .

to be spin-coated at a film thickness below 200 nm. The use of this diluted Olin for 266 nm LIL has already been described elsewhere, e.g. Murillo et al [126]. Although the resist is not very stable after dilution, here we applied this resist system for multi-exposure experiments, producing a mesoscale quasi-regular nanoarray (for an overview of nanoarray definitions see once more figure 1). Subsequently, a process for nanogratings by a single exposure into positive chemical amplified resist, PEK500 (Sumitomo Chemical) was established for a systematic evaluation of scaling line width, pattern period (pitch) and pattern depth (aspect ratio). To study the interaction of biological material (tissue, blood, proteins) with these nanostructured surfaces, experiments have been initiated in collaboration with Jansen's group at Radboud University Nijmegen Medical Centre, the Netherlands. First, systematically scaled large-area nanogratings were fabricated by solvent cast replication from the large-area  $(2 \times 2 \text{ cm}^2)$ master achieved by the LIL process and subsequent silicon etching. Secondly, these nanostructured surfaces were investigated for their cell response. The full details of large-area nanolithographic master fabrication using LIL and anisotropic dry etching will be communicated elsewhere [133]. Finally, using a third type of resist, the negative acting MA-N2403 (Microresist), some experimental results are shown that demonstrate the potential of using high-contrast resists during LIL-multi-exposures to generate integrated micro-nanoarrays. The full process details including the utilization of a variety of these structures in biological systems are to be published shortly. Here, we only stress the multitude of processing possibilities when a LIL nanolithography system is at hand.

6.1.2. Results. Results of the LIL nanofabrication process are shown in this section. Multiple exposure experiments at two different angles of incidence (14° and 15.4°) and repetition of these exposures after a substrate rotation of 90° were carried out to achieve a latent nanopattern grey-scale image in the OiR photoresist, here acting as a low-contrast resist at  $\lambda = 266$  nm to generate Moiré type features. Subsequently, the resist was developed in a single step in an immersion bath for 1 min using a standard OPD4262 positive resist developer. Figure 8 shows these structures evolved from interference of the multiple set of exposures.

Figure 9 demonstrates the potential of p-CAR pattern transfer by parallel plate reactive ion etching of a dot pattern into a layer of silicon nitride aiming at a pattern period (pitch) of 380 nm, which refers to an angle of incidence of  $20.48^{\circ}$  in the two orthogonal exposures. The dots can be subsequently coated with magnetic, conducting material. Previously, these types of nanoarrays were fabricated using an orthogonal exposure in OiR resist. Subsequently, the samples were used in a method for field-emission distance control by le Febre *et al* [127]. Applying the improved LIL process utilizing p-CAR resist, as shown here, will further support well controlled sample preparation for PM recording experiments.

Figure 10 shows the nanograting pattern fidelity of the line width/space ratio at six different points across the diameter of a 4 inches silicon wafer measured by scanning electron microscopy. This result demonstrates the issue of controlling the uniformity of such continuous nanoarrays over a large manufacturing area. This aspect remains a challenge for the exposure tool designer in selecting suitable optics.

When replacing the low-contrast OiR resist with a highcontrast negative resist, e.g. here MA-N2403 (Microresist) a

![](_page_13_Picture_2.jpeg)

**Figure 9.** Dot arrays on silicon. Orthogonal exposure at 20.4° angle of incidence each. The scanning electron micrography shows a cross-sectional view of the dots etched in silicon nitride. The p-CAR resist mask has not yet been removed.

highly defined integrated micro-nanoarray pattern results by the same type of multi-exposure carried out in the previous experiment. A result of such micro-nanoarray using the negative resist is presented in figure 11.

6.1.3. LIL nanoarrays for cell response studies have also been investigated. It has been demonstrated that biological processes are activated when a material exposes a specific nanotopology [134, 135]. Inspiring also our work, for example, cultured fibroblast on PMMA surface that were nanostructured using AAO as a template in nanoimprinting were investigated by Kim *et al* as mentioned before [80]. Recent work by our collaborator showed that e-beam lithography can be applied to manufacture a template containing nanopatterned fields of  $500 \times 500 \,\mu\text{m}^2$  that are reproduced in polysterene and subsequently tested for their cell response. The lines and spaces nanopattern within each field had various pitches but always at a line width-to-space ratio of 1:1 [136]. Original work relates back to micropatterned surfaces for cell alignment in cell culture [137]. Here, the size and geometrical dependence of biological response is systematically evaluated in cell culture experiments dedicated to the improvement of tooth implants. LIL can be used in this context for two specific reasons: its capability for largearea nanoarray formation as well as its potential to be used for a simple and cheap maskless exposure onto non-flat surfaces. So far, large-area nanoarrays with a diversity of line width, pattern period and depth have been investigated for their fibroblast and osteoblast response and will be reported shortly. Initial results on cell response to artificial nanotopology utilizing LIL have been presented [138]. The large-area patterning supported by the LIL technique is of particular importance in gaining statistically relevant results from the cell culture proliferation experiments. Substrates produced by e-beam lithography are often too small in surface area and hardly allow more than a few cells attaching to one specific pattern.

# 7. Conclusion

In lithographic processes the control of the transfer of a layout first into a masking layer (resist) and sequently in a functional material sometimes with multiple steps allow the creation of devices with an enormous variety of capabilities. Of course, still the electronics domain is at the frontier of lithographic techniques reaching the level of features at the scale of a few nanometres. Other disciplines caught up quickly and even bring about their own lithographic techniques. A broad choice of techniques exists. Only a few techniques will be selected in the next decade to lead electronics market innovation. In the leading electronics industry EUV is probably preferred above techniques which are currently exploring multiple e-beam/ion-beam sources. In research investigating nanoscale behaviour as well as in specific (e.g. PMs for magnetic data storage) and niche manufacturing (e.g. optical devices) templated techniques are gaining significance. Specifically, NIL, gradually replacing the mainstream UV-photolithography, gets strong. When it comes to nanoarrays, IL became a valuable addition to the range of techniques. The ability of chemical and biological substrates to coordinate ordering at the molecular scale is also about to reach the level of control required in industry, however, not yet on the wafer-scale as we are used to in commercial applications. In respect to further deepen our understanding of controlling device functions at nanoscale dimensions, research in the field of nanolithography will stay extremely diverse and may be guided mainly by a technique at hand than actually a choice by design. It is, therefore, of particular importance to develop new cost-to-lithographic performance models, which give the designer of novel devices some insight into this diversity. Yet, giving the designers a choice between either a well established but limited process line, e.g. as it is found in microelectronics complementary metal-oxide semiconductors (CMOSs), or a novel lithography that capitalizes on the advantage of delivering very unique devices properties is not an easy one. The multitude of parameters involved, when it comes to establishing a production line, makes such a choice difficult. Realizing that the classical approach to lithography is in the domain of a plane, there are next to direct-write techniques, e.g. using a focused ion beams, also other techniques being developed. The extension to the out-of-plane dimension may be either supported by stacking (assembly), multilayer processing, folding, or three-dimensional lithographic techniques such as two-photon lithography [139]. However, so far only few technologies have shown true potential for three-dimensional crystal lattices of several magnitudes of lengthscale, e.g. holographic lithography [140]. Whether these meta-materials find real applications beyond the example of photonic crystals is not yet foreseen. Patterning of nanoarrays, e.g. for multilayers useful to perpendicular magnetic recording, x-ray diffraction, EUV diffusers and SAW devices, as well as surface modification by nanostructuring of implant materials, however, is further vividly investigated in our laboratory.

![](_page_14_Figure_2.jpeg)

**Figure 10.** Single exposure at  $40.6^{\circ}$  angle of incidence [133]. The scanning electron micrographs are taken on the middle line of the wafer to demonstrate the pattern distortion due to optical aberrations. However the useable area of high uniformity still stretches across a few cm<sup>2</sup> during the full 4 inch wafer-scale exposure process. Areas of high pattern fidelity vary with the angle of incidence applied and the total mirror surface area being exposed uniformly by the source.

![](_page_14_Figure_4.jpeg)

**Figure 11.** Nanohole-microarray formed by high-contrast negative resist LIL-multi-exposure. Courtesy of van Wolferen, University of Twente, 2008. Details of this process are disclosed.

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